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HYDROGEN-OXYGEN ELECTROLYTIC REGENERATIVE FUEL CELLS

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National Aeronautics and Space Administration
Lewis Research Center
21000 Brookpark Road
Cleveland, Ohio
Attn: D. G. Soltis

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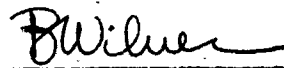
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Prepared by


M. Klein

Approved by:


E. Findl
Manager
Chemical and Fluid Systems Dept.


B. M. Wilner
Acting Manager
Energy Conversion Div.

ELECTRO-OPTICAL SYSTEMS, INC.

Pasadena, California

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1. INTRODUCTION

This report reviews the progress made on the development of a regenerative hydrogen-oxygen fuel cell under NASA Contract 3-2781, during the period 1 July, 1964 to 31 December 1964. The program objective is the development of an electrically regenerative hydrogen-oxygen fuel cell that will be superior in performance to currently available rechargeable batteries. The device under development consists of a cell stack that is utilized both as an electrolyzer during charge periods and as a fuel cell during discharge periods. Integral gas storage tanks are used to contain the hydrogen and oxygen gas generated during charge. Such a device offers advantages in the area of watt hours per lb., high ambient temperature operation, (plus the ability to be sterilized) and greater cycle life than can be obtained from existing secondary batteries.

A two phase program is being conducted. Phase I consisted of the design, development and testing of a nominal 75 watt, 44 watt-hour, 6 cell unit to demonstrate the feasibility of a multi-cell regenerative device. During Phase I, emphasis was placed on developing useful, reliable component designs and experimentation to determine if unforeseen chemical and electrochemical difficulties might develop using these components. The major objective of Phase II is the design and development of a 500-1000 watt, 600 watt-hour, 34 cell, minimized weight unit, for evaluation as a flight prototype.

2. SUMMARY

Design problems uncovered during the first half of 1964, were thoroughly evaluated and eliminated during this report period. [A single cell unit and two six cell units were fabricated during this time. The single cell unit was built to establish optimum design characteristics and to evaluate the effect of changes in such controllable variables as the asbestos matrix (type, compression, and electrolyte content), temperature, charge rate, discharge rate and electrode catalyst loading.] As a result of careful redesign and single cell testing, the 75 watt, 6 cell unit was successfully cycled over an extended time period. Performance levels achieved, exceeded the nominal output levels. Subsequent cycling on the same unit indicated that levels > 90 watts could be obtained, and that by increasing the operating temperature from 70°C to $\approx 125^{\circ}\text{C}$, outputs approaching 120 watts are obtainable.

The primary factors resulting in the greatly improved performance and reliability were as follows:

1. Increased electrode catalyst loading from 10 mg/cm² to 20 mg/cm².
2. Improved bipolar plate design.
3. Use of fuel cell grade asbestos instead of electrolytic or commercial grades.
4. Improved volume compensation bellows design.

Performance data and details of the single and multi-cell designs are presented within the report.

3. TECHNICAL DISCUSSION

As a result of thermochemical reaction problems encountered during the previous semi-annual report period, a thorough review and analysis of the original design and operating parameters was made. This review resulted in establishing certain areas where new information was required and where new design principles had to be employed. The following sections discuss in detail the information obtained, the designs arrived at, and the test results obtained using this data.

3.1 Asbestos Studies

There are three major criteria governing the use of porous asbestos as the separator material in the regenerative fuel cell. First, it must have low porosity to H_2 and O_2 gases, second it must exhibit high electrolyte absorbtivity, and third it must have structural and chemical stability. Three types of asbestos have been investigated to determine their ability to meet the first criterion. These types were (a) commercial grade (b) electrolytic grade and (c) fuel cell grade. Commercial asbestos is high in impurity content, exhibits high porosity and wide variability in absorbtivity. It has good structural stability however. The electrolytic grade has intermediate properties as to purity, and the fuel cell grade represents the highest purity material available in bulk.

In order to provide some measure of the relative porosity of the various grades and the effects of certain variables on this porosity, a series of leakage rate tests have been conducted. Certain of these tests and the test procedure have been reported previously.⁽¹⁾ For clarity in this presentation, we have included data from reference 1 where pertinent to the discussion. It must be emphasized that the data presented herein is for comparative purposes only, and is not meant to be used for permeability computations on an absolute basis.

One of the factors governing asbestos leakage rate is the degree to which it must be compressed to prevent gas leakage. This leakage can occur both through and around the edge of the asbestos matrix. Low degrees of compression permit the storage of relatively large amounts of water, but the resulting matrix is more susceptible to gas leakage. The reverse is true for high degrees of compression. In order to evaluate the effect of compression, a series of gas leakage tests was conducted.

Data obtained with commercial grade asbestos are shown on Figure 1. Compression ratios, i.e., the initial/final asbestos thickness, were varied between $7/6$ and $7/3$. Initial asbestos thickness, i.e., prior to compression was ~ 0.070 inches. The type of curve presented on Figure 1 is similar to that presented in Figure 2 showing the effect of electrolyte/asbestos weight ratios on leakage rate. It is interesting to note that the $7/4$ compression ratio appears to be the optimum for commercial grade. One might expect the $7/3$ ratio to show less leakage, but apparently, at the $7/3$ value, an appreciable quantity of the electrolyte is squeezed out of the asbestos, increasing the permeability somewhat over the $7/4$ value.

In addition to compression ratio tests, additional leakage rate experiments were carried out using electrolytic and fuel cell grade asbestos. Results of these and data from reference 1 on the commercial grade are shown on Figures 3, 4 and 5. With the exception of the 0.015" thick fuel cell grade, the leakage rates of both electrolytic and fuel cell grades are much lower than that exhibited by commercial asbestos. Logically, the leakage rate could be expected to be an inverse function of the thickness. However, the data indicates definite anomalies in this regard. There are a number of possible explanations for these deviations from logic such as (a) variations in processing involved to obtain individual thicknesses, (b) batch to batch differences, i.e., non-reproducibility of the asbestos (c) uncontrolled experimental parameters and (d) a combination of the first three. Since the test procedure is so simple, and in the majority of

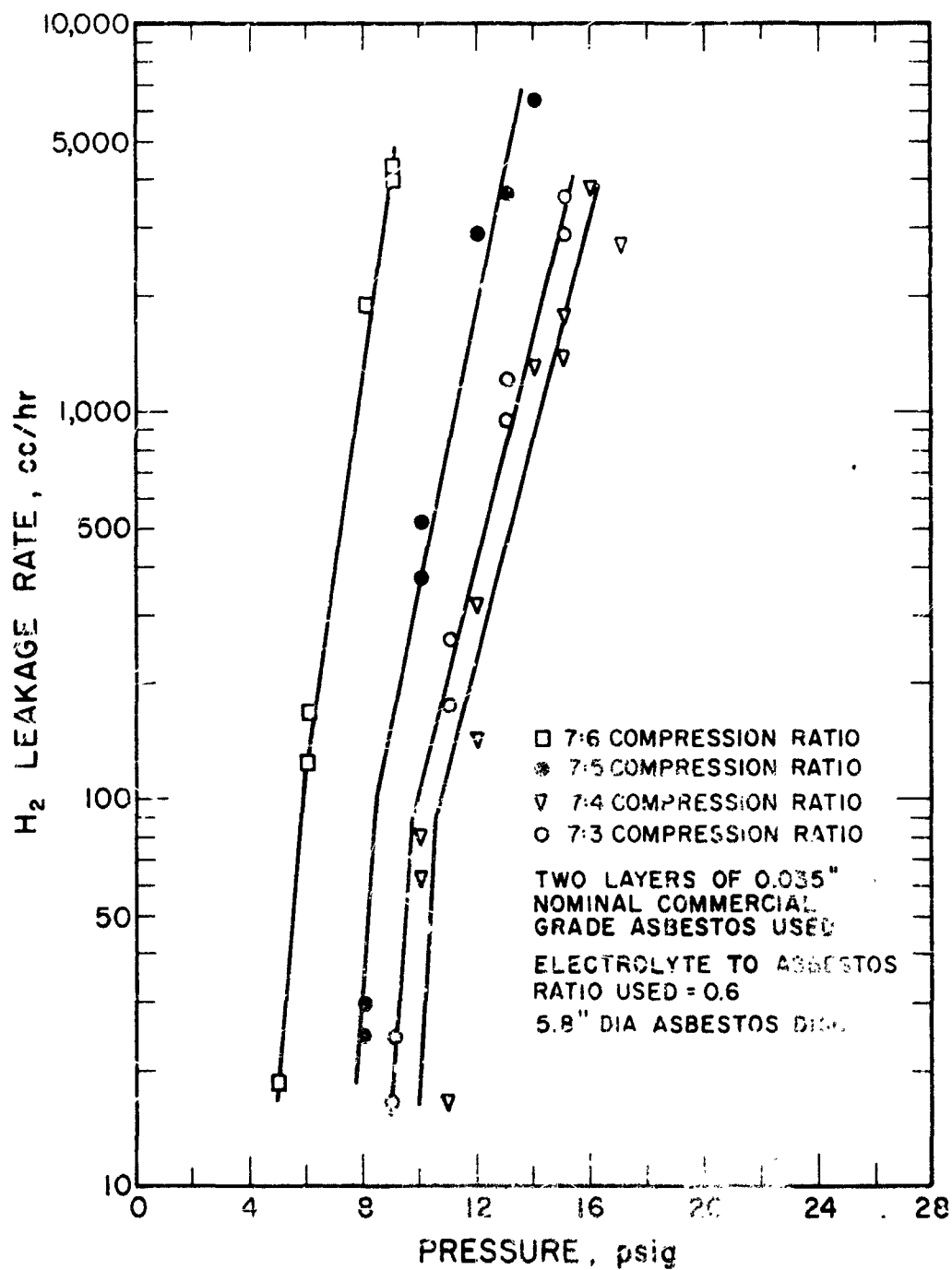


FIG. 1 EFFECT OF ASBESTOS COMPRESSION RATIO ON PERMEABILITY OF HYDROGEN THROUGH TWO LAYERS OF 0.035 INCH COMMERCIAL ASBESTOS

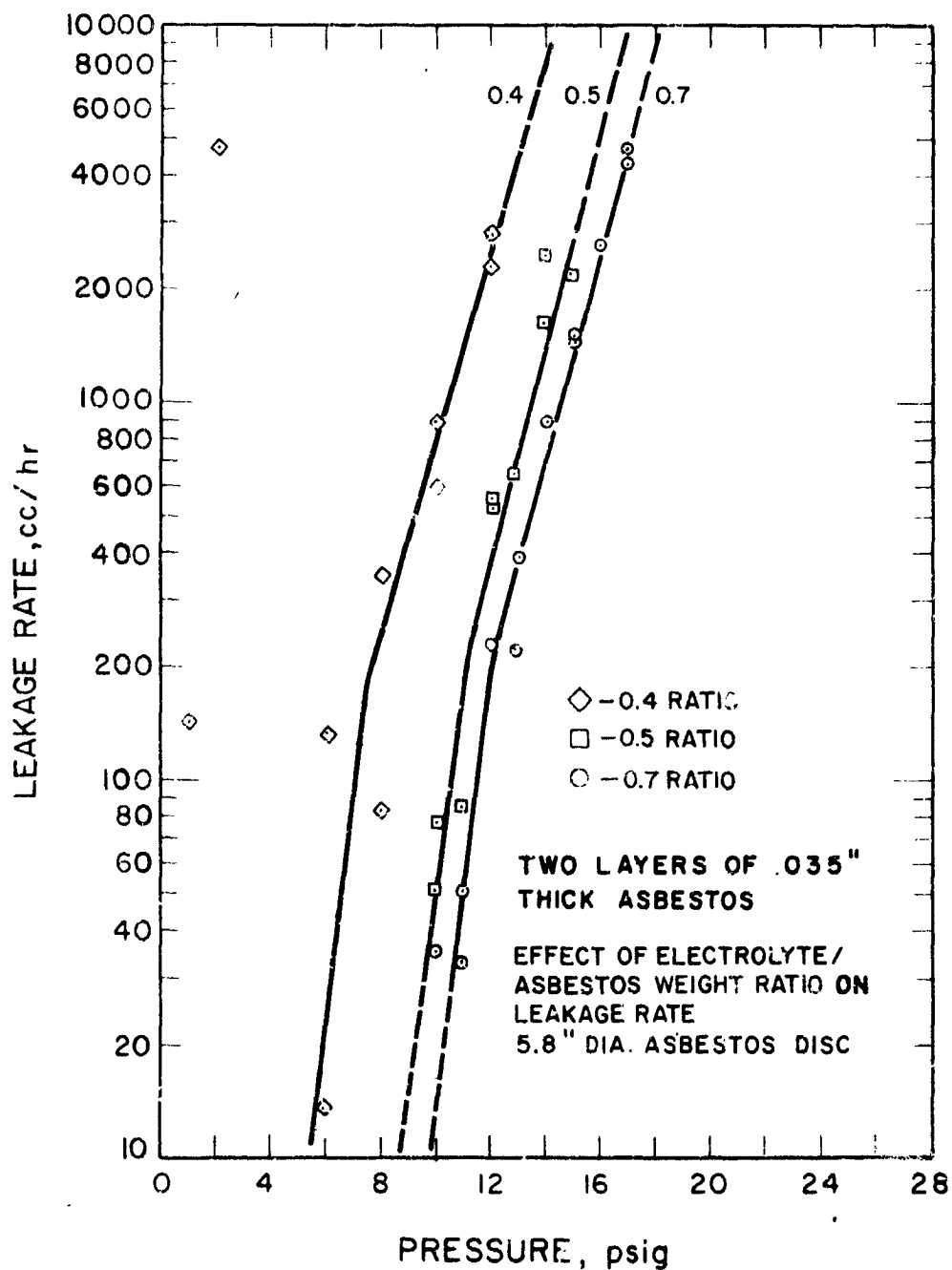


FIG. 2 EFFECT OF ELECTROLYTE CONTENT ON LEAKAGE RATE OF HYDROGEN THROUGH 0.035 INCH COMMERCIAL ASBESTOS

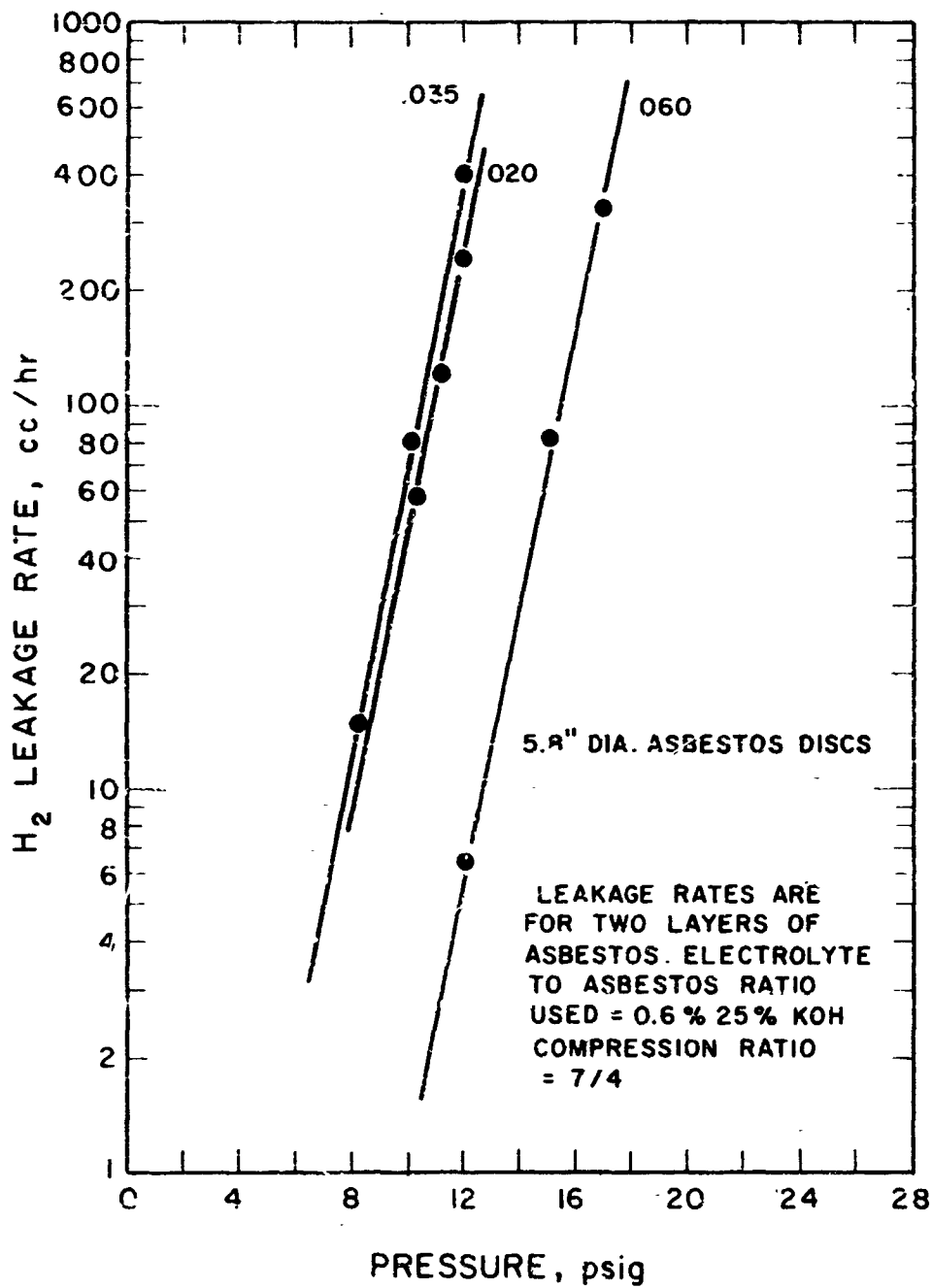


FIG. 3 HYDROGEN LEAKAGE RATE FOR COMMERCIAL GRADE ASBESTOS

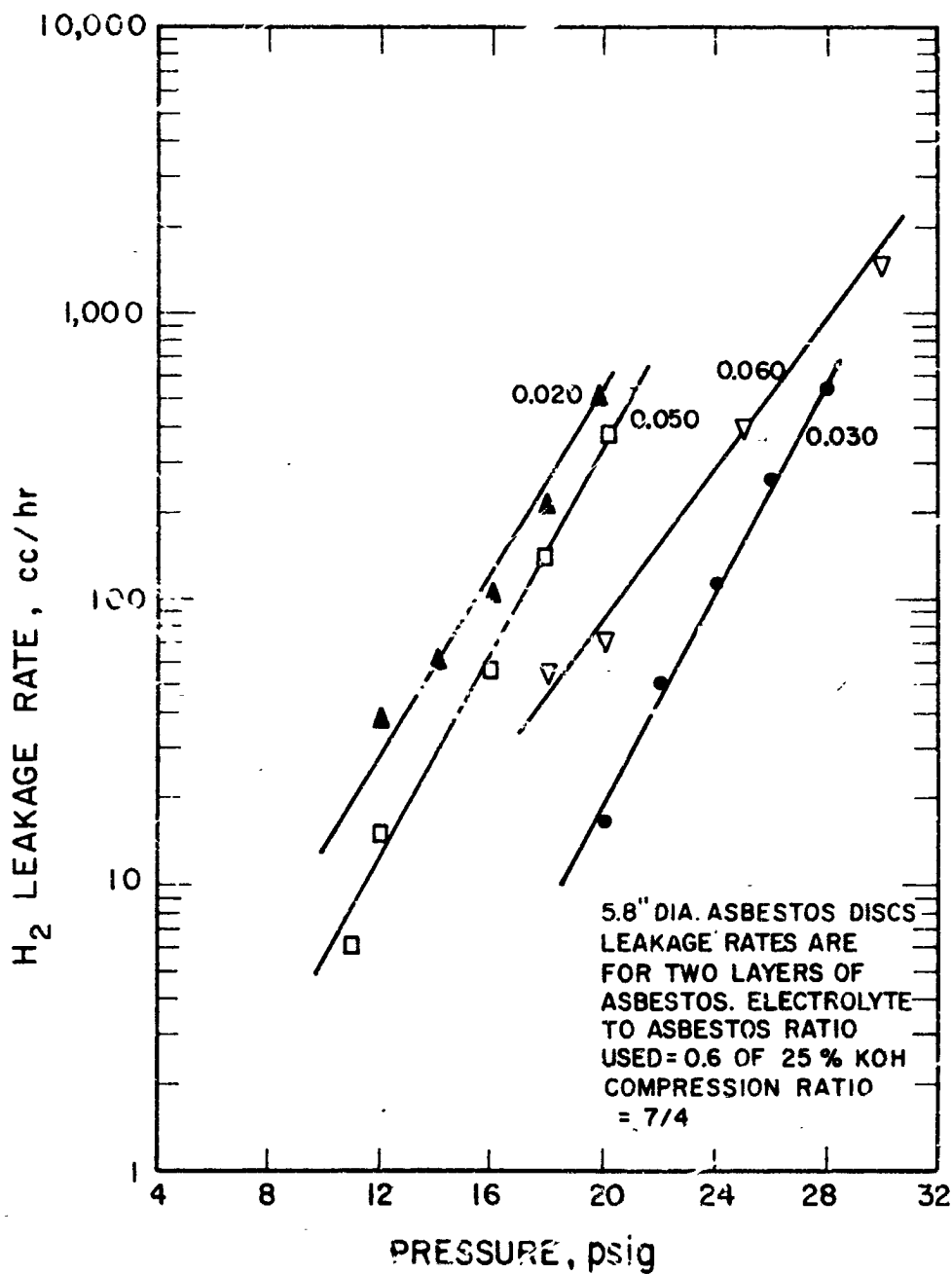


FIG. 4 HYDROGEN LEAKAGE RATE FOR ELECTROLYTIC GRADE ASBESTOS

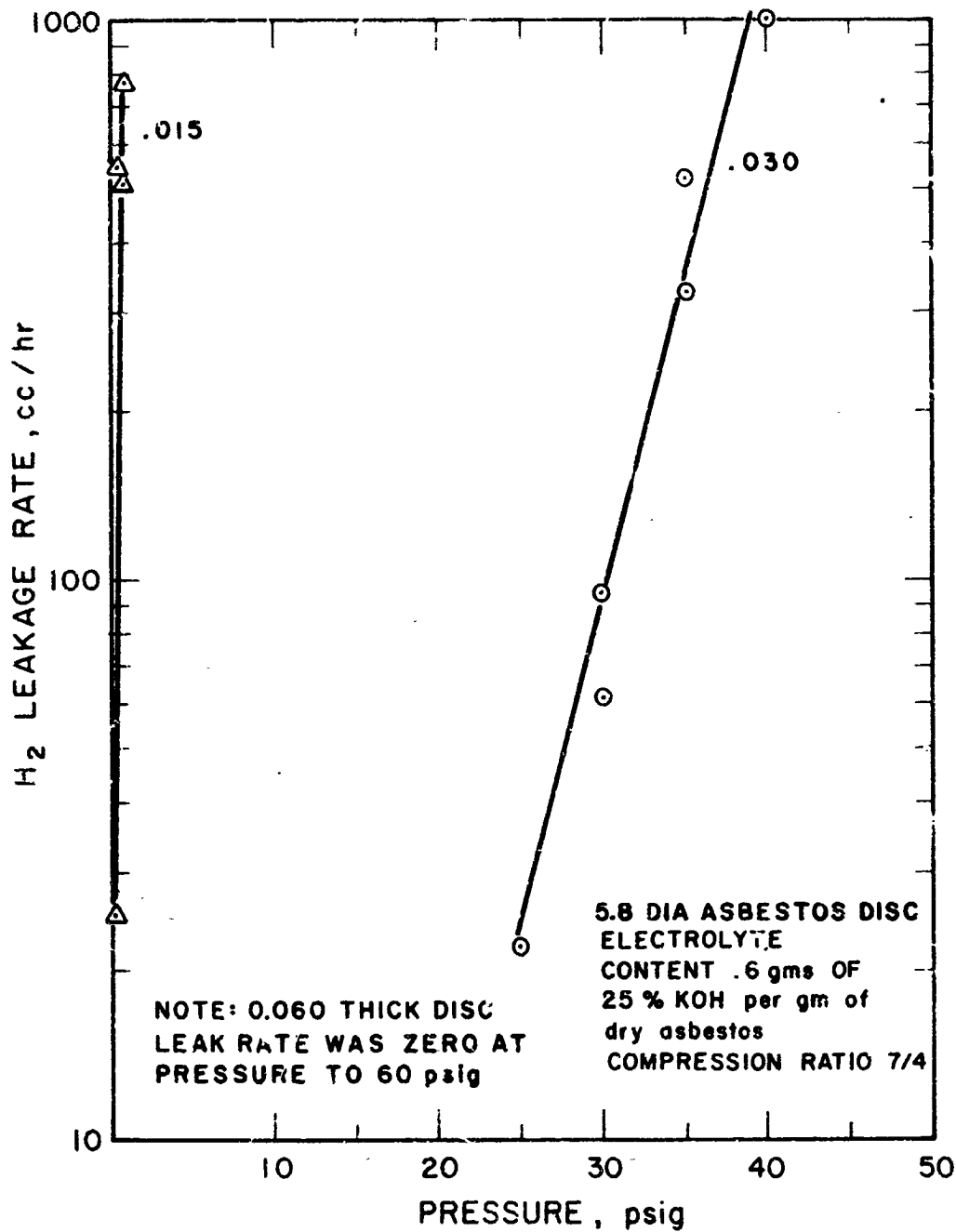


FIG. 5 HYDROGEN LEAKAGE RATE FOR FUEL CELL GRADE ASBESTOS

the tests fairly reproducible, we feel the anomalies are an inherent problem of the asbestos manufacturing process. This is emphasized by the data of Figure 6, showing the effect of multiple layers of fuel cell grade asbestos on leak rate. Here, one would expect leakage rates through multiple layers of equal total thickness to be nearly equal or perhaps have lower leakage rates for more layers of material. However, this is not indicated.

Regardless of the leakage rates shown, the important parameter as far as leakage is concerned is the pressure at which leakage initially occurs. We call this point the breakover pressure. It varies considerably between grades of asbestos, but the trend is as follows:

<u>Grade</u>	<u>Breakover Pressure-Psig</u>
Commercial	6-12
Electrolytic	10-20
Fuel Cell	20-40 (other than 0.015" thick)

Variations in compression and electrolyte content will, of course, shift these values somewhat. However, it is evident that relatively large differentials can be sustained across the various types of asbestos, and that the fuel cell grade is superior to the other two.

3.2 Design Details of the 75 Watt Unit

The present design contains certain changes from the design previously evaluated and reported in (1). These changes were made to improve reliability and performance and are listed below.

1. Replacement of the rubber pressure balancing diaphragm by a stainless steel bellows.
2. Incorporation of an improved bipolar plate containing an integral gas port seal.
3. Incorporation of improved gas distribution by elimination of bipolar plate external drill holes.
4. Elimination of thermal storage tanks.
5. Incorporation of unipotential tankage.

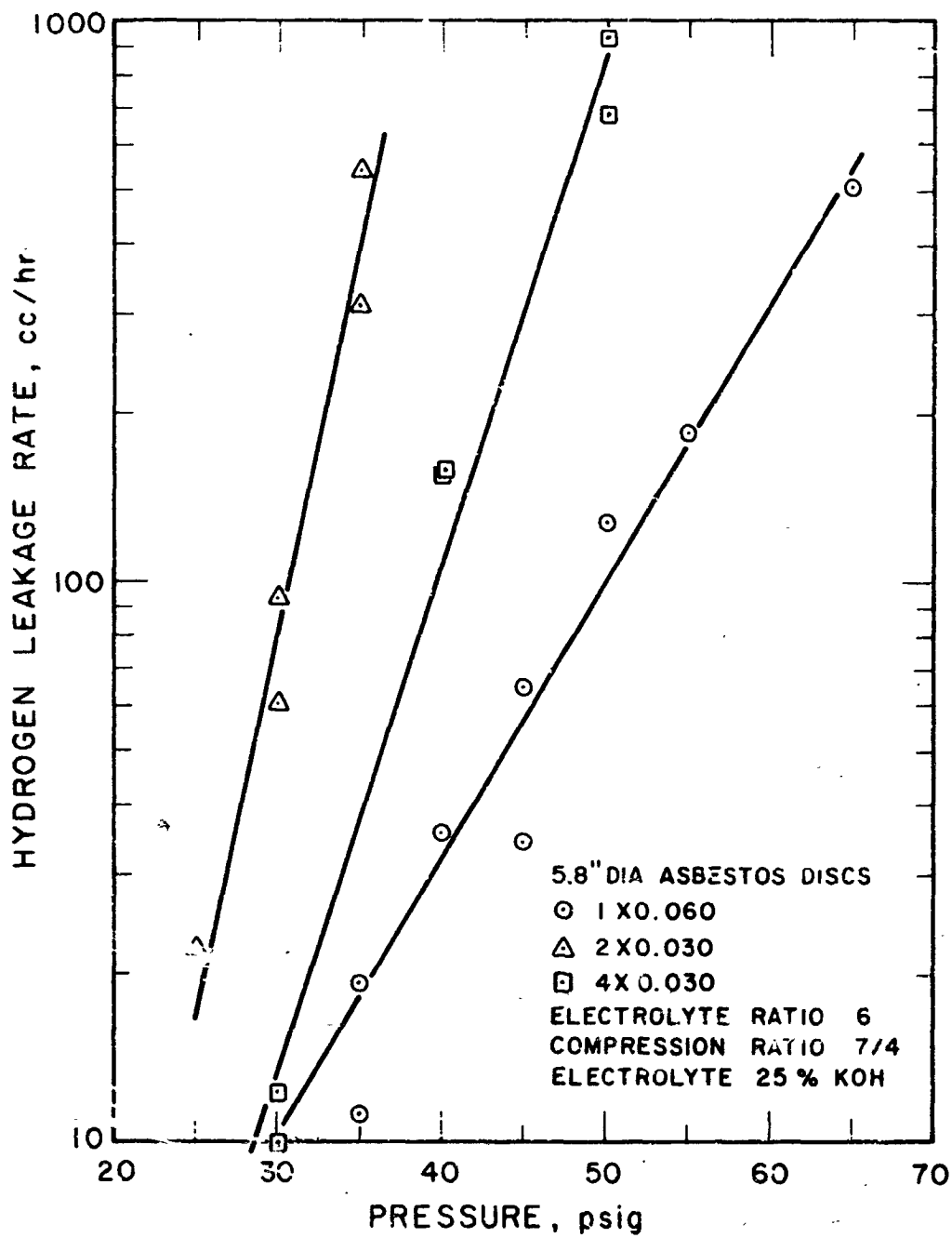


FIG. 6 HYDROGEN LEAKAGE RATE FOR FUEL CELL GRADE ASBESTOS

Each of these changes has been made along with other modifications to simplify fabrication and assembly, increase gas porting area to the pressure balancing bellows, and to minimize the hazards involved in testing. Figure 7 shows a photo of the assembled 75 watt unit and Figure 8 shows an assembly drawing of the unit giving details of internal construction.

As in the previous 75 watt model, six series connected cells using 6-inch diameter electrodes have been employed. However, the electrodes presently being used contain 20 mg/cm^2 of catalyst rather than 10 mg/cm^2 as used previously. Two types of catalyst are being employed, namely 100 percent platinum and 50 percent platinum/50 percent palladium. The latter type are used interchangeably with the former as oxygen electrodes with no apparent change in catalytic activity. Fuel cell millboard asbestos has been used exclusively in all 75 watt assembly tests. The mats are ≈ 50 mils thick, weigh 22 grams, and are compressed to 40 mils in the cell stack. Twenty-nine grams of 40 percent KOH are added to the mat as the electrolyte.

Assembly is accomplished by stacking the individual bi-polar plates containing the electrodes and asbestos mats. A stainless steel bellows, which acts as a volume compensation device, is attached to the oxygen end plate. This volume compensation is required for two purposes, first, to correct any errors in volume due to design or machining errors, and second to compensate for non stoichiometric evolution of H_2 and O_2 during the initial portions of the charge and discharge cycle. Non stoichiometric evolution results from electrochemical oxidation to NiO of uncatalyzed portions of the nickel plaque electrode in contact with the electrolyte. On discharge, this nickel oxide, amounting to approximately 30 amp. minutes equivalent, is reduced prior to the consumption of gaseous oxygen. (Section 3.6 discusses this further.)

Once the cell stack, including the bellows is assembled, the negative lead from the high pressure feed through is attached to the

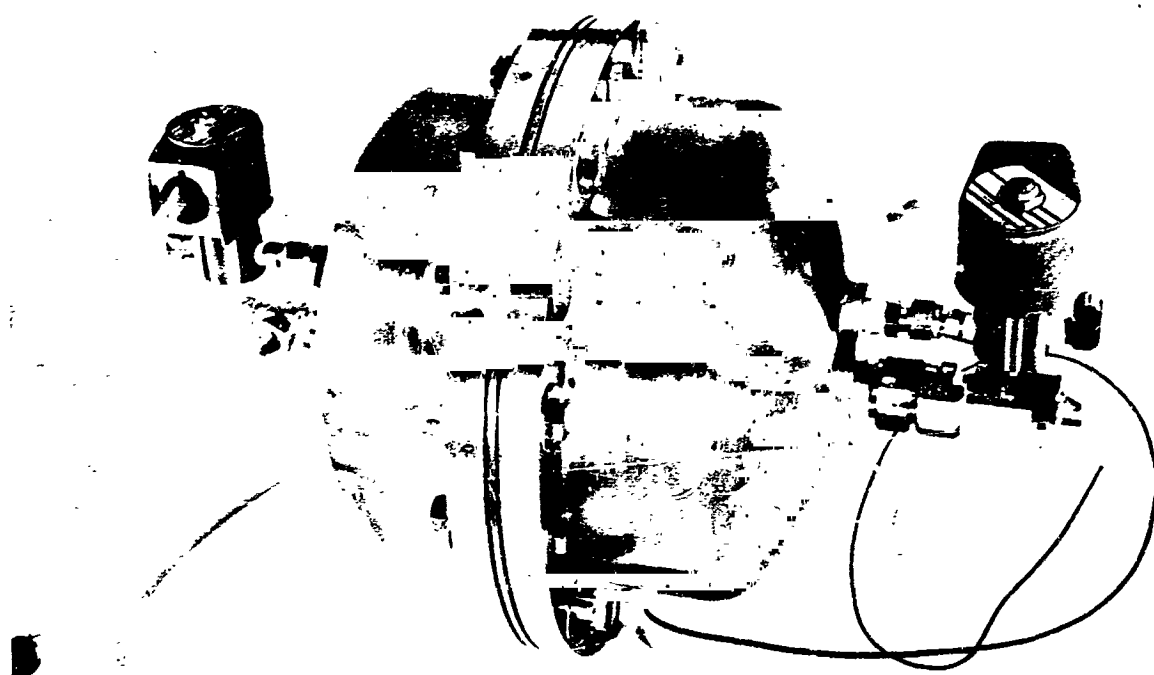


FIG. 7 75-WATT FUEL CELL ASSEMBLY

stack, along with an iron-constantan thermocouple. The integral tanks are then installed and bolted externally. In order to simplify tank construction, both the H_2 and O_2 tanks were made identical. Volume adjustment was made by installing two teflon spacers, one of which was also used as a bellows extension stop.

3.3 Single Cell Design

In order to evaluate certain variables affecting performance, a single cell, 6-inch electrode diameter fuel cell was designed and fabricated. This cell contains features similar to that used on the multi-cell unit, but does not contain a pressure balancing mechanism. Therefore, the H_2 and O_2 tank volumes within the cell had to be carefully balanced prior to its use in the regenerative mode of operation.

The single cell unit consists of two circular plates of monel containing gas cavities in an approximate 2-1 volume ratio. Machined into these cavities are grooves for electrode back-up plates and electrodes of the same, i.e., 6-inch diameter, that are utilized in the multi-cell unit. The two halves are bolted together by means of a series of bolts along the periphery. Sealing is accomplished by the use of "O" rings. A glass reinforced epoxy spacer is employed between the two plates to provide electrical insulation between the positive and negative sides of the cell. The insulating spacer thus provides the spacing between the two plates and also fixes the compression on the asbestos matrix. Since the diameter of the asbestos matrix is somewhat larger than the 6-inch electrodes, the peripheral edge of the matrix is compressed against the cell halves inside the insulating spacer, effectively preventing leakage around the asbestos. Assembly is accomplished by stacking one end plate, back up plate, electrode, asbestos mat, second electrode, and second back-up plate, and then bolting the entire assembly together.

Certain fittings, valves, and instrumentation are attached to each of the chambers for pressure measurement and for flushing the unit. Major cell components were fabricated out of monel to minimize

corrosion problems. Since the cell does not contain a volume balancing mechanism to assure a 2-1 volume ratio, it was necessary to adjust the volumes by adding external tubing to the cell. The entire cell was set up in an oven that contained lead-ins for electrical and flushing gas connections. Figure 9 shows a photo of the assembled single cell and Figure 10 shows an assembly drawing of the unit giving details of the internal construction.

3.4 Instrumentation

For both single and multi-cell tests, readout and control instrumentation has been assembled in one test cabinet for consolidation and ease of handling. The test unit now consists of a load bank with variable resistors to control discharge current, a power supply to supply charging current, a power supply for the two pressure transducers, and two vacuum tube volt meters for total and differential pressure readout. A recycling timer is also included which automatically switches the cell from charge to discharge. (It is presently set for a 65 minute charge and a 35 minute discharge cycle.) The unit also contains outputs to a multipoint recorder for recording cell voltage, cell discharge current, and total cell pressure. Cell temperatures are also recorded on a multi-point recorder, using thermocouples located internally and on the external periphery of the cell. Differential pressure is recorded on a continuous strip chart recorder.

3.5 Test Facility Construction

Due to the possible hazards involved in testing the multi-cell unit, a new test facility was designed and built with corporate funds. The facility is essentially a block house containing the steel test chamber used during the preceding stages of the program. Modifications to the steel chamber improving its pressure venting characteristics were also included. The block house is attached to the fuel cell laboratory and all test operations are carried out by remote control. Suitable control panels and visual monitoring facilities have been provided.

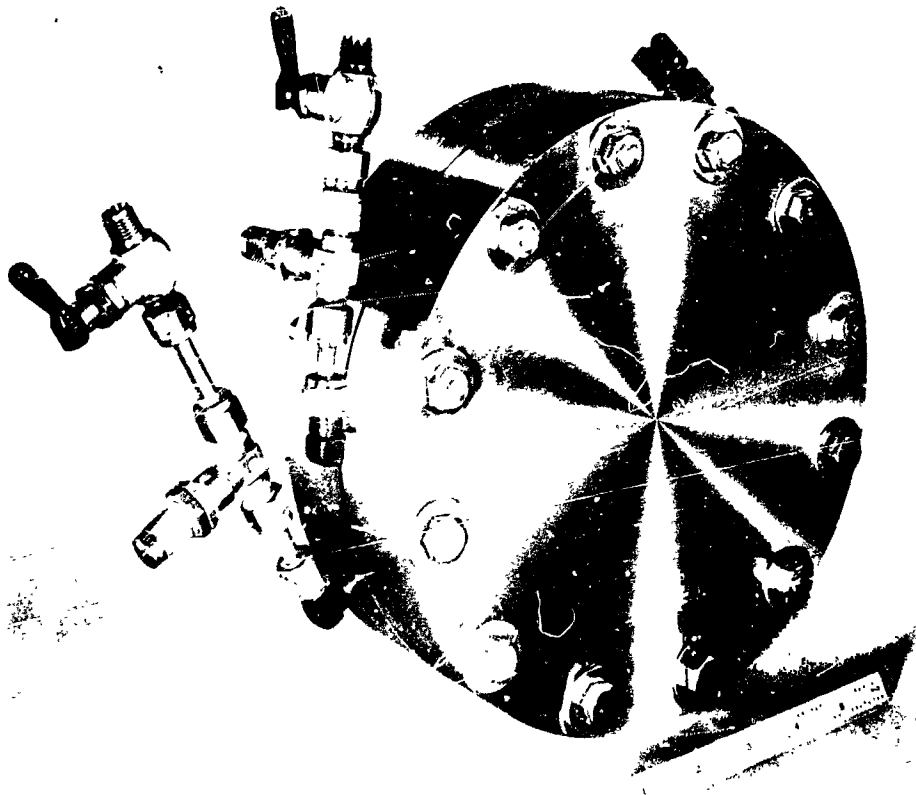
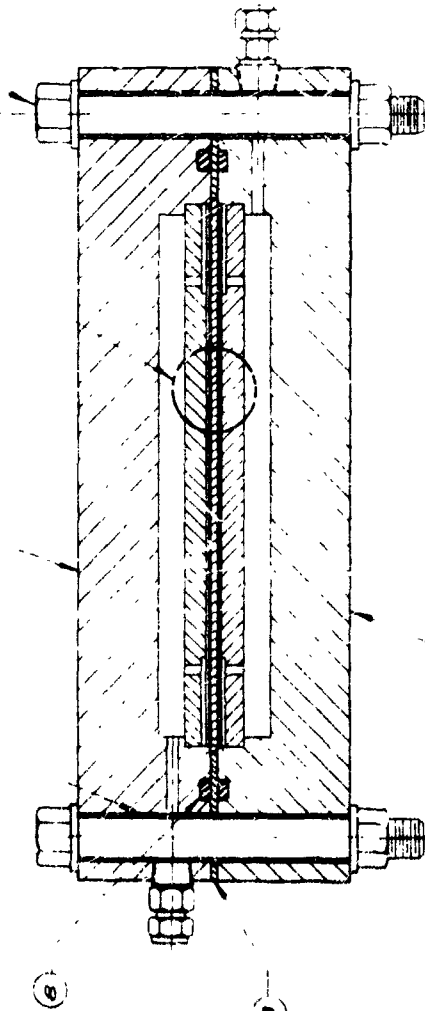


FIG. 9 SINGLE CELL ASSEMBLY

SEE DETAIL A

910111



DETAIL A
SCALE: 2X



QTY		PART NO.		DESCRIPTION		MATERIAL		REMARKS	
1	12	12	12	SEPARATOR	3910	ADBESTOS			
2	11	11	11	WASHER-FLAT	3910	CRES			
12	10	10	10	NUT-HEX	3910	CRES			
12	9	9	9	SEAL-W-HEX	2444	CRES			
2	8	8	8	PARMER	4505	NEOPRENE			
12	7	7	7	O-RING	6110	PHENOLIC			
2	6	6	6	BOLT INSULATOR	6110				
2	5	5	5	ELECTRODE	6110	NICKEL			
2	4	4	4	ELECTRODE	6110	CRES			
1	3	3	3	INSULATOR	6110	GLASS FIBER			
1	2	2	2	INSULATOR	6110	CRES			
1	1	1	1	INSULATOR	6110	CRES			

UNLESS OTHERWISE NOTED: 1. DIMENSIONS IN INCHES 2. DECIMAL FRACTIONS 3. ANGLES 4. RADIUS 5. DIA.		APPROVED DATE		APPROVED DATE		APPROVED DATE	
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FIG. 10

3.6 75-Watt Cell Testing

After initial assembly and check out, the redesigned 6-cell unit was installed and tested in the new fuel cell testing facility. Appropriate feed-throughs for electrical and pneumatic connections have been installed in the test facility, and were connected to the cell. The unit was flushed 10 times by pressurizing with hydrogen and oxygen to 50 psig and then venting the gases. The ambient temperature in the test oven was then raised to 70°C, and the cell was put on cycle. The cycle consisted of 65 minutes charge at 9.6 amps, and a discharge load of 0.33 ohms set to give 75-80 watts during discharge. In the initial cycles, the performance of the unit looked stable and satisfactory. Therefore, it was decided to continue cycling and conduct a continuous 48-hour test which represented a major test objective. Data for cell voltage, cell pressure, and temperature during the continuous 48-hour test is shown in Figures 11a--11e. Throughout the entire testing period, the differential pressure swing did not rise more than 0.2-0.3 psi. When the cell pressure reached 380-390 psi, the charger was automatically shut off, and the cell allowed to sit on open circuit until the cycling timer (set at a 65 minutes charge duration) switched the unit to discharge.

After completion of the 48-hour test, the cell was vented and allowed to sit at temperature on open circuit. In the following days, a number of other charge-discharge cycles were conducted to determine various performance capabilities, and to see if any deterioration in performance would occur. Figure 12 shows a series of discharge curves at different temperatures and loads. As can be seen, at a slightly higher temperature, the unit was capable of discharging continuously at 25 amps.

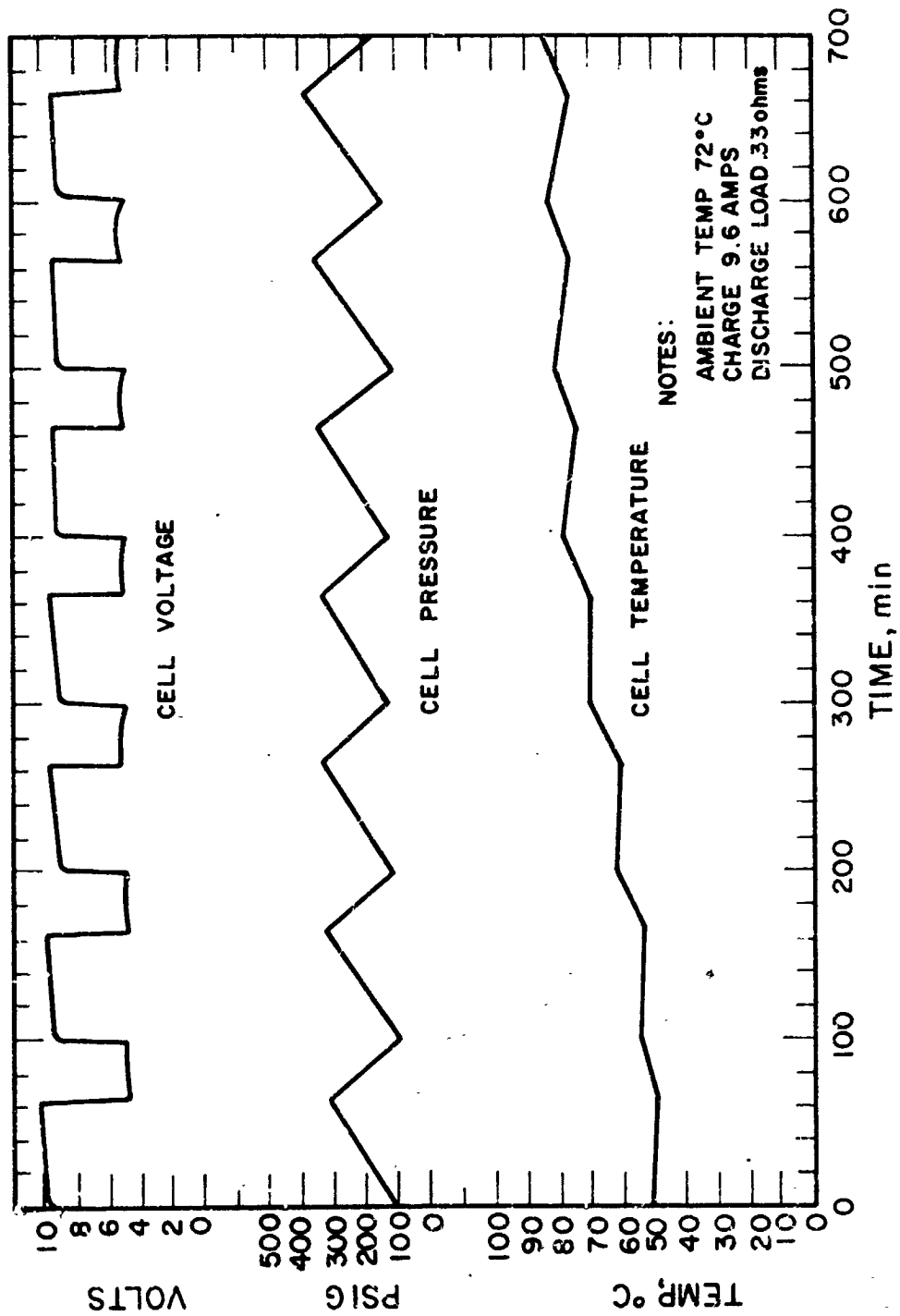


FIG. 11A CONTINUOUS 48-HOUR CYCLING OF 75-WATT ELECTROLYTIC REGENERATIVE $H_2 - O_2$ FUEL CELL

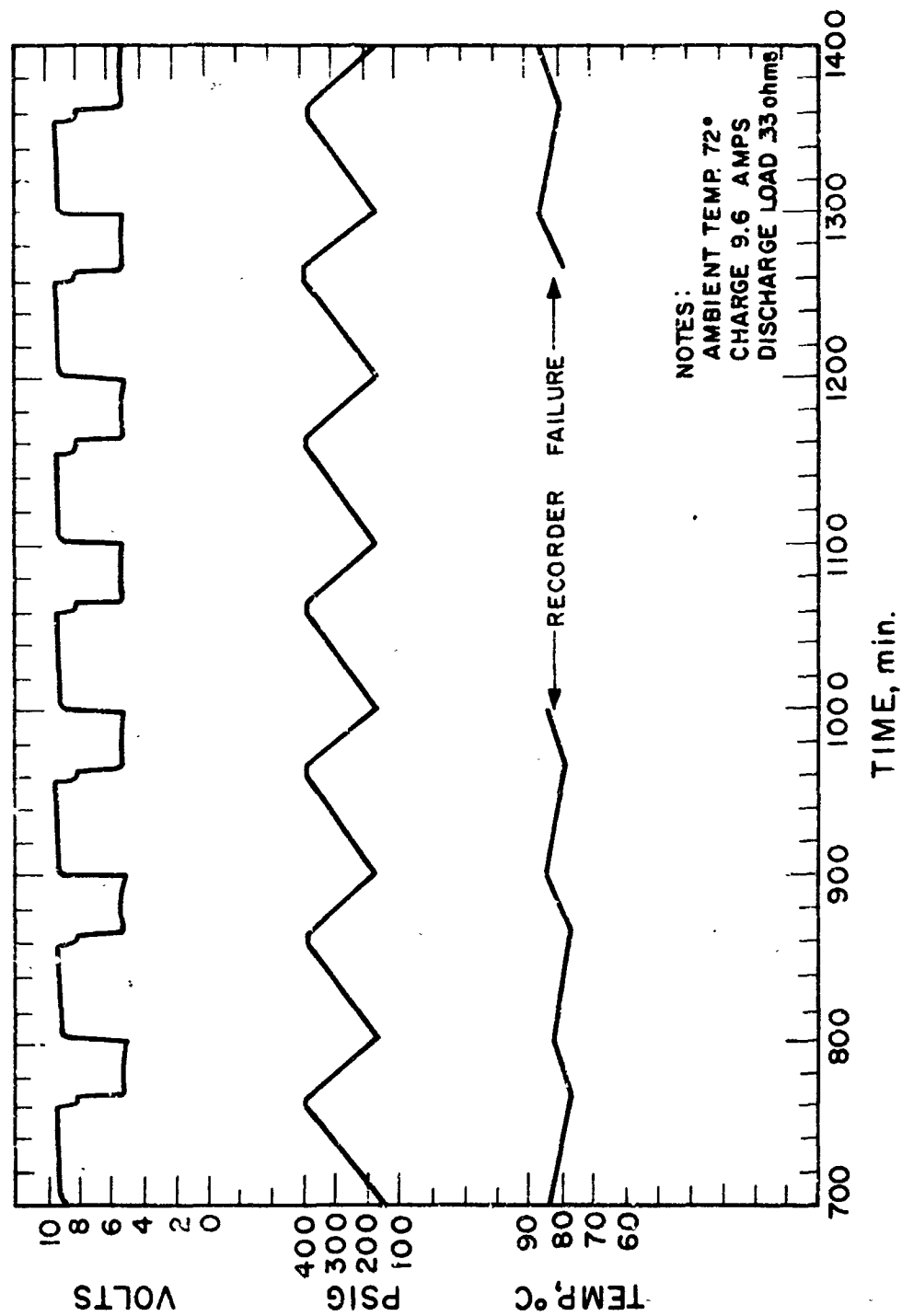


FIG. 11B CONTINUOUS 48-HOUR CYCLING OF 73-WATT ELECTROLYTIC REGENERATIVE H_2-O_2 FUEL CELL

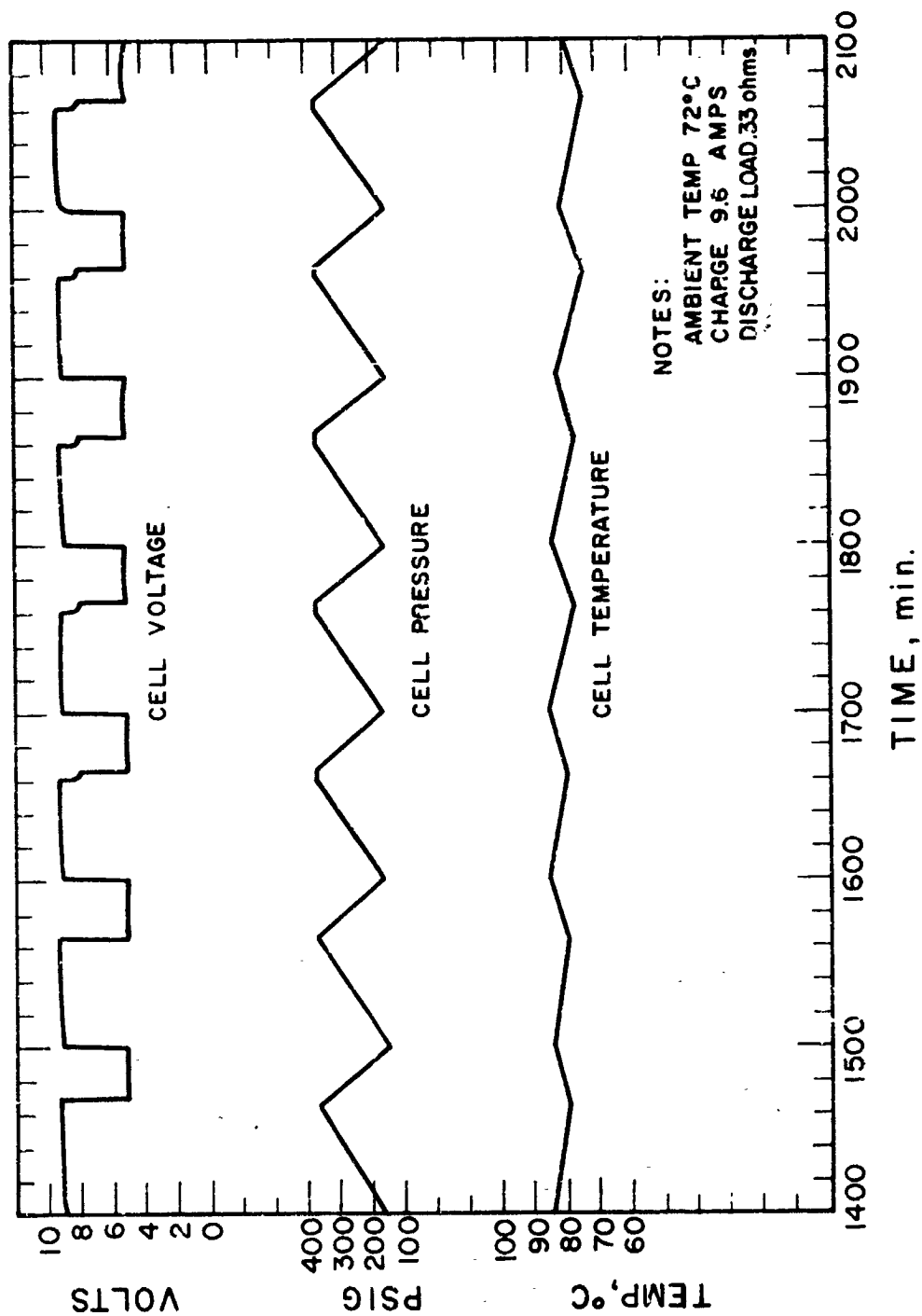


FIG. 11C CONTINUOUS 48-HOUR CYCLING OF 75-WATT ELECTROLYTIC REGENERATIVE H_2-O_2 FUEL CELL

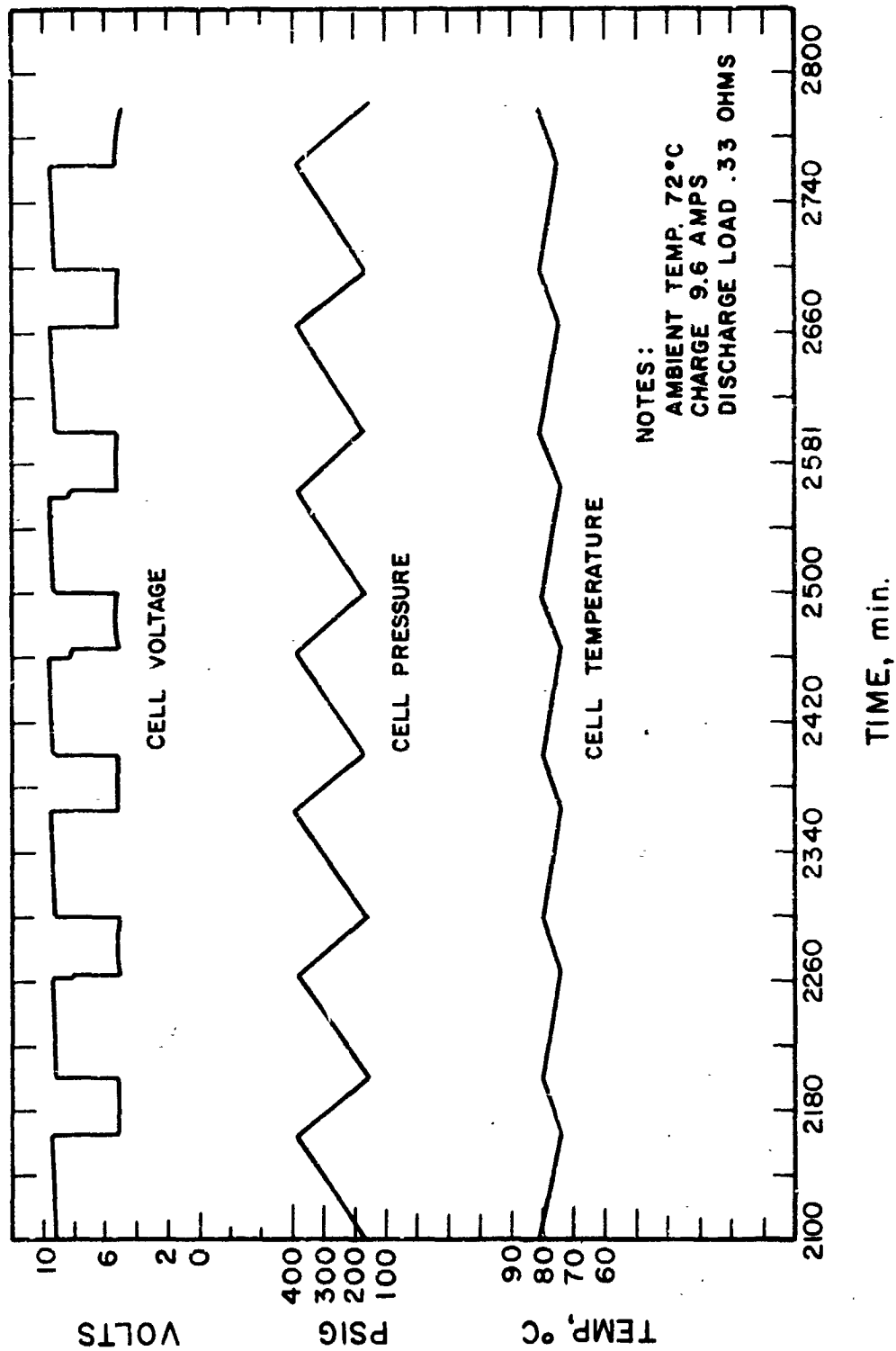


FIG. 11D CONTINUOUS 48-HOUR CYCLING OF 75-WATT ELECTROLYTIC REGENERATIVE H_2-O_2 FUEL CELL

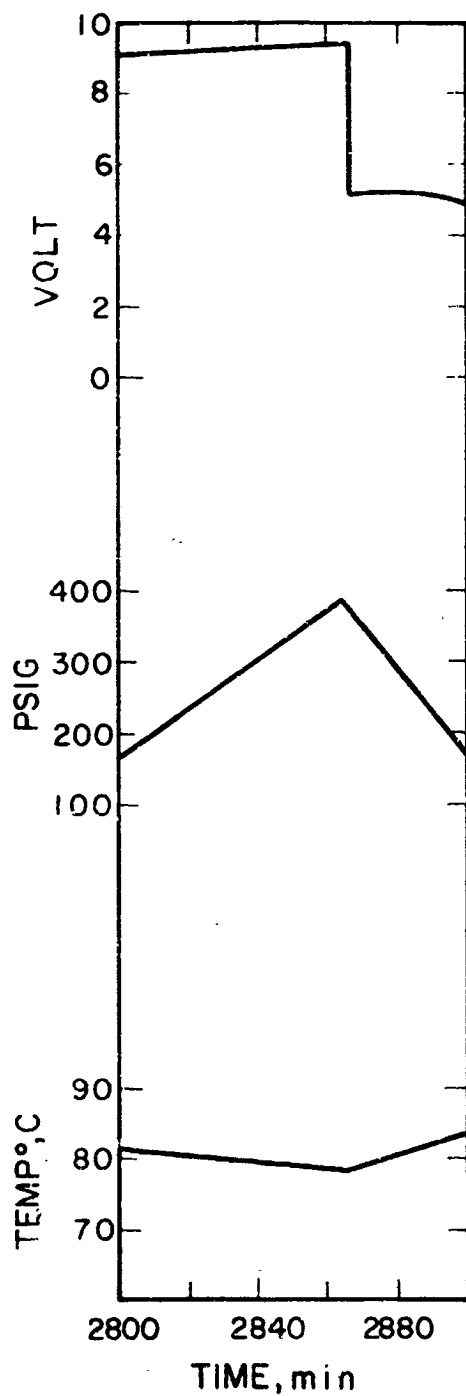


FIG. 11E CONTINUOUS 48-HOUR CYCLING
OF 75-WATT ELECTROLYTIC
REGENERATIVE H_2 - O_2 FUEL
CELL

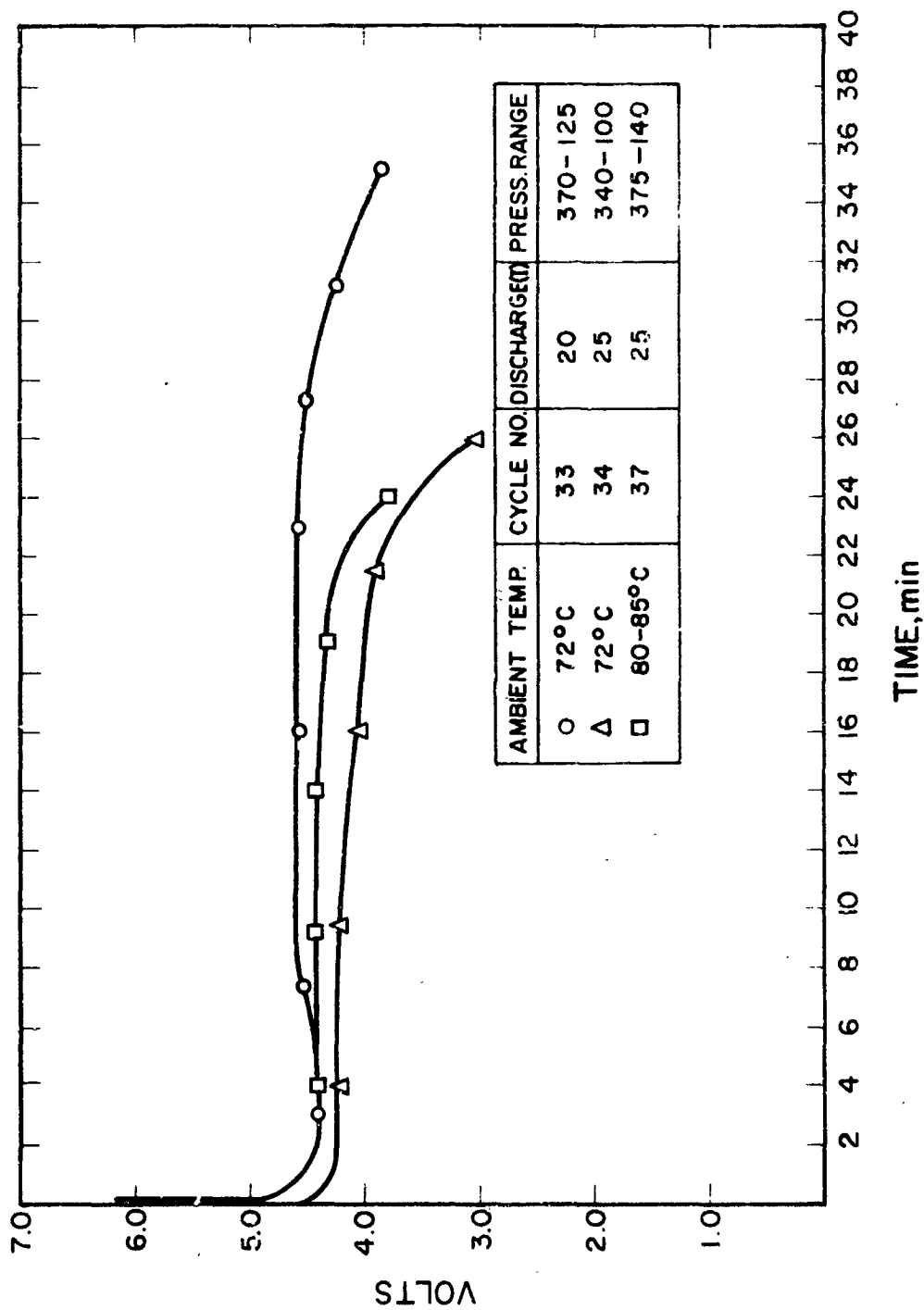


FIG. 12 DISCHARGE DATA 75-WATT ELECTROLYTIC REGENERATIVE H_2-O_2 FUEL CELL

Figure 13 shows a test of cell capacity. The unit was charged until the cell pressure reached 450 psi, and then continually discharged at 15 amps until the cell voltage reached 3 volts. (It should be noted that at the end of charge period, the cell voltage was still relatively low, indicating that the mats contained additional water that could have been utilized for additional capacity.) Figure 14 shows a voltage vs. current curve for the unit, and the accompanying power output curve for this test. After an accumulation of a total of 39 cycles, the unit exhibited slightly deteriorated performance on both charge and discharge. The unit was then disassembled and examined. The individual mats had gray to black discoloration adjacent to the hydrogen electrode and the viton rubber on the mat edges had turned from green to black. Electrolyte squeezed from samples of two of the cells showed that the concentration had dropped to approximately 13 percent KOH from the 40 percent level. This drop in % KOH is discussed in Section 3.8.

A second multi-cell unit was later assembled using components identical with the first with the following exceptions; (a) the oxygen electrodes consisted of a mixed catalyst, i.e., 10 mg. of platinum, and 10 mg. of palladium per sq. cm., (b) insulating washers used on stack bolts were fabricated from glass epoxy rather than the previously used nylon in order to prevent relaxing of the washers during load at elevated temperatures, and (c) the mats contained no edge sealant.

The unit was cycled in the normal manner, i.e., 35 minute discharge, 65 minute charge. During the first two cycles, the temperature was gradually raised, and the unit discharged at approximately 15 amps. at 5.2 volts. For the remaining 14 cycles, performance increased slightly, and the discharge current was set at approximately 18 amps. The unit was cycled only during an 8 hour day. Overnight the unit was kept at elevated temperature. Prior to shut down, the unit was vented of all pressurized gas, and the following morning was repressurized to the final pressure level of the previous evening. Figure 15 shows a typical charge-discharge curve of the cell during the cycling period.

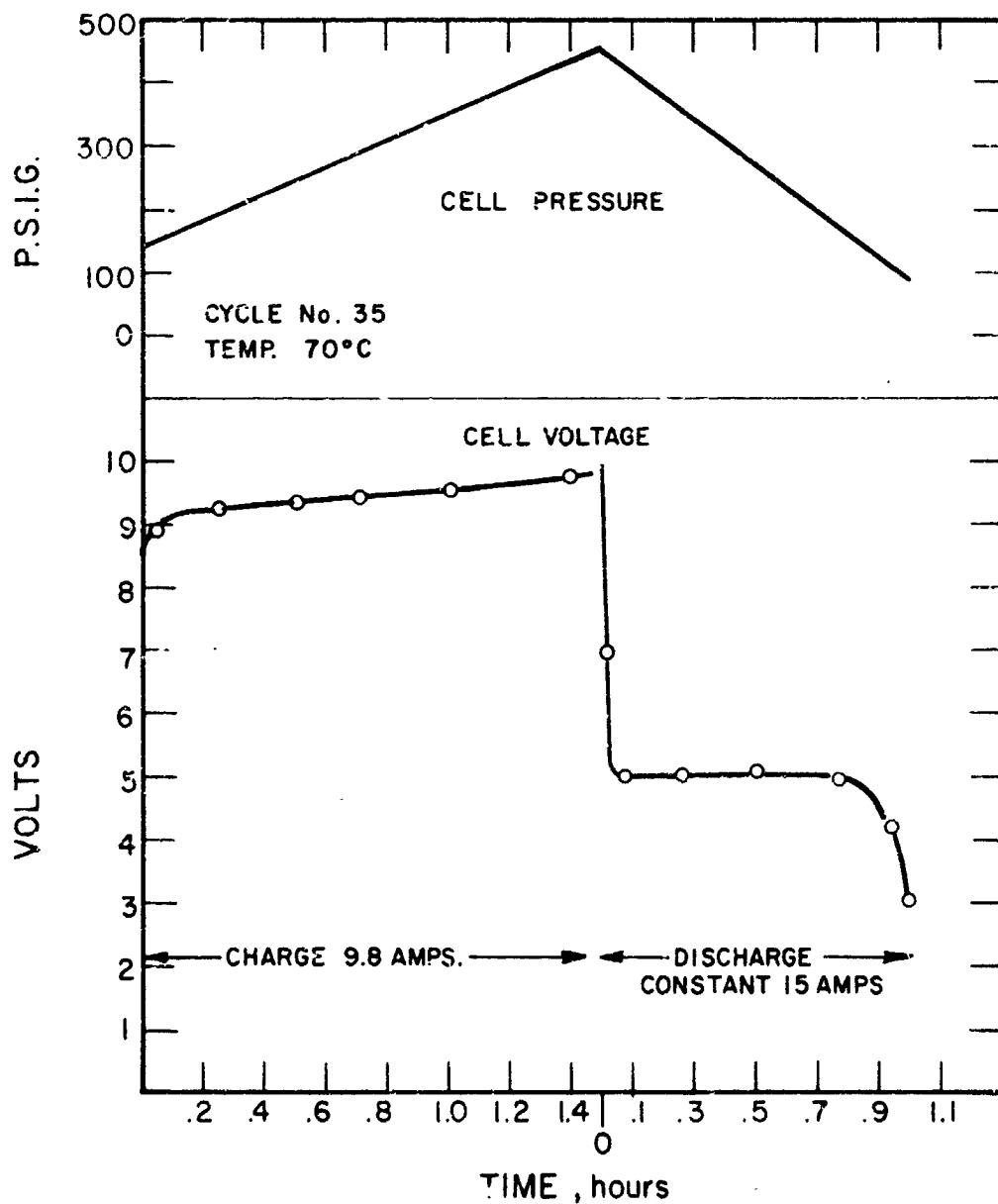


FIG. 13 CAPACITY TEST CYCLING DATA H_2 - O_2 ELECTROLYTIC REGENERATIVE FUEL CELL

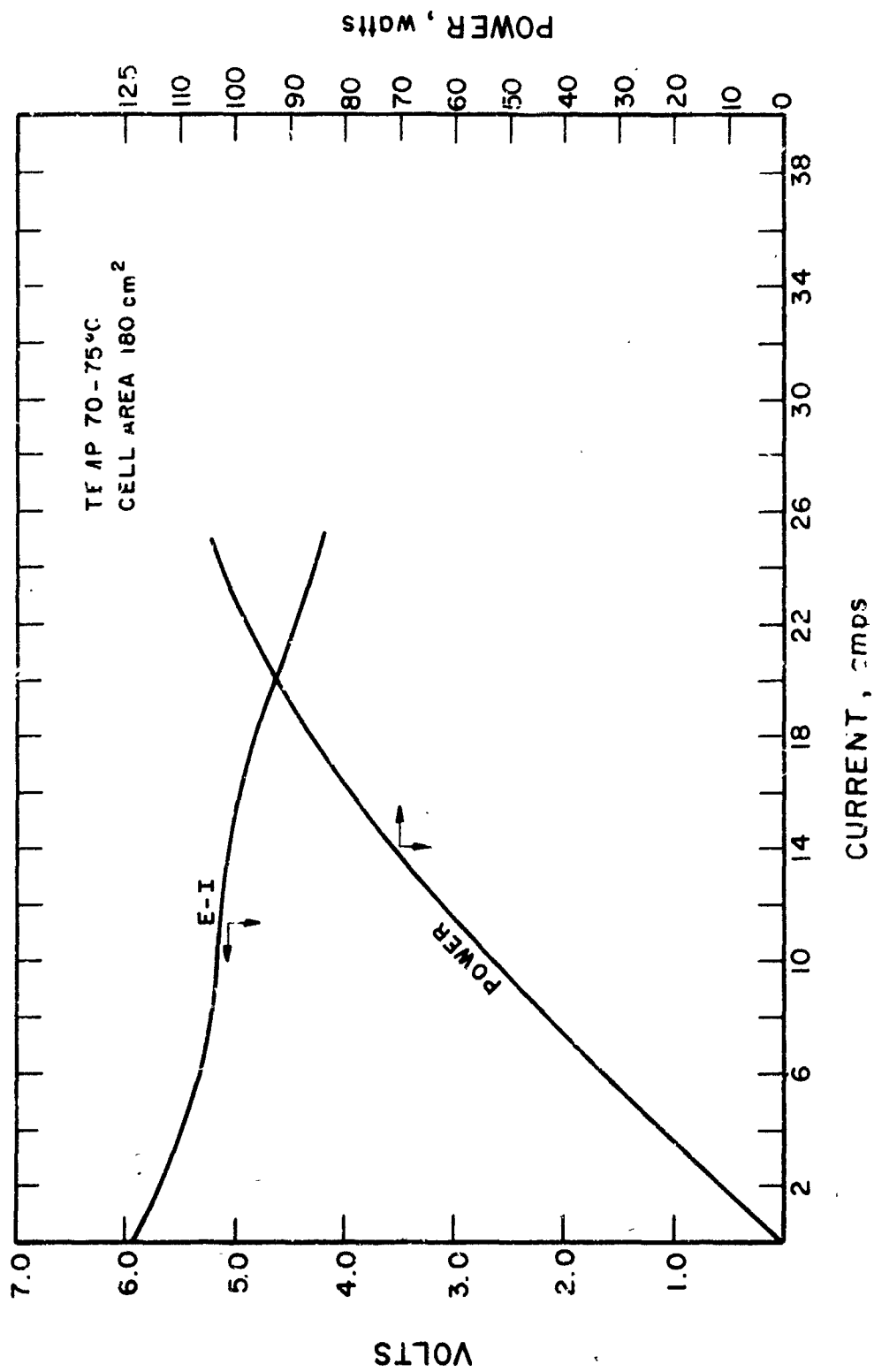


FIG. 14 PERFORMANCE DATA 5-WATT (6 Cell) ELECTROLYTIC REGENERATIVE H_2-O_2 FUEL CELL

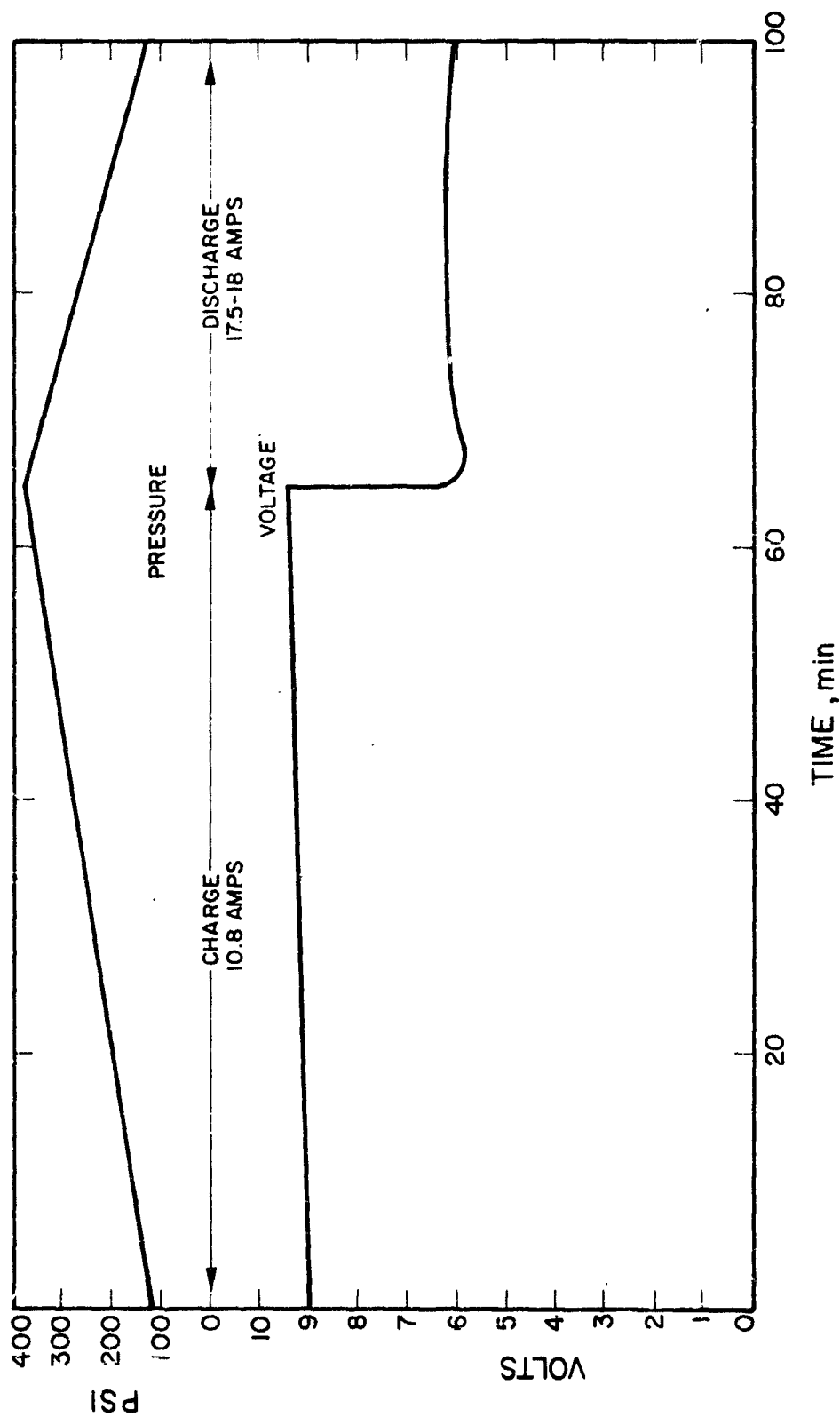


FIG. 15 H_2-O_2 REGENERATIVE FUEL CELL CYCLE PERFORMANCE OF SIX CELL UNIT S/N 102

On the morning following the 16th cycle, when the cell was being pressurized to be put into service, it was noted that there was cross leakage between gas compartments, and the cell was not capable of holding a differential pressure. In addition, a slight rise in temperature was recorded internally in the cell (15° - 20°) during this filling period. The unit was therefore disassembled and examined.

Examination of the disassembled unit revealed that the stack bolts had relaxed considerably. These bolts were initially torqued to 75 inch/pounds. Inspection of the unit revealed that the bolts were essentially at 0 torque after the testing period. A relaxation of this type most probably released the compression on the mats, and allowed gas leakage between cell compartments at the mat periphery. Considering this difficulty, it has been decided to utilize some form of compression loading springs on the stack bolts to eliminate the relaxation problem in the future. The mats had a grey discoloration adjacent to the hydrogen electrode, but otherwise were satisfactory, and they did not stick to the electrodes within the entire assembly. Analysis of electrolyte concentration, within two of the mats of the disassembled cell, revealed concentrations of 33 and 34 percent KOH.

3.7 Single Cell Tests

During this report period, a number of single cells were assembled and subjected to various tests. These tests are described in Table I which summarizes the features and results obtained. The initial assemblies were used mainly to debug the test set up, and to balance cell volumes. In the initial tests, preliminary cycling was conducted at room temperature. The oven was then turned on and the temperature was allowed to rise and equilibrate at 70°C . Internal leakage difficulties were encountered in initial testing, as indicated by poor Faradaic efficiencies. Examination of the cells after disassembly revealed that the electrodes had collapsed into the back-up plate slots which are used as gas feeds. This resulted in a lack of asbestos matrix compression at

TABLE 1
SUMMARY OF SINGLE CELL TESTS

Cell #	O ₂ Electrode # # Catalyst	H ₂ Electrode # # Catalyst	Mat Thickness Inches and Grade	Mat Dry Wt.	KOH Wt.	Comments	Results
1	2 10 Mg Pt/ 1 cm ²	1 10 Mg Pt/ 1 cm ²	0.050- 0.020 Electrolytic	30.3	25.4	18.2	Used cell to balance volumes and check out test circuit. Cycled cell 7 times. Stopped test due to excessive internal leakage.
2	4 "	3 "	0.050- 0.020 Electrolytic	29.0	25.4	20.3	In initial test differential transducer failed and cell had to be reassembled
3	4 "	3 "	0.050- Electrolytic	29.9	25.4	20.9	Did not cycle cell due to inability to hold pressure differential across mat.
4	4 "	3 "	0.050- 0.020 Electrolytic	30.2	25.4	21.1	Cycled 4 times. Discontinued due to internal leakage
5	4 "	2 "	0.050- 0.020 Electrolytic	30.9	25.4	21.6	Cycled 3 times. Discontinued due to internal and external leakage.
6	4 "	3 "	0.050- 0.020 Electrolytic	30.0	25.4	21.0	Placed Spacers behind gas feed ports.

Table 1 (con.)

7	1	10 mg Pt/ 2 cm ²	2	10 mg Pt/ 2 cm ²	0.020- 0.030- 0.020 Electrolytic	31.6 25.4 23.3	Cycled 12 times. Stopped test due to low discharge voltage on 10th, 11th, and 12th cycle. Possibly caused by gas leakage and cell drying out.
8	1	"	2	"	0.020- 0.020- 0.020 Electrolytic	27.0 25.4 23.3	Cycled cell 4 times. Stopped test due to low discharge voltage.
9	6	"	5	"	0.020- 0.020- 0.020 Electrolytic	27 25.4 23.3	Cycled 7 times, left on stand 3 days at temp. After stand exhibited low voltage on discharge.
10	4	"	3	"	Commercial Grade 0.035- 0.035 Electrolytic	31 25.4 24.3	Cell test stopped due to excessive internal leakage.
11	8	"	7	"	0.035- 0.004- 0.035 Commercial Grade	34 25.4 28	Cell had excessive internal resistance
12	1	"	2	"	0.020- 0.030- 0.020 Electrolytic	31.8 25.4 23.5	Cycled 2 times left on stand at temp. 2 days. Gave degraded performance.

Table 1 (con.)

13	6	10 mg Pt/ 2 cm ²	5	10 mg Pt/ 2 cm ²	0.020- 0.030- 0.020 Pure asbestos fuel cell millboard	29.1	25.4	23.3	Added water to the cell by pressurizing and discharging.	Cycled 14 times. No. deterioration noted but cell had low discharge voltage.
14	-	10 mg Pt/ 2 cm ² + 10 mg Pd cm ²		20 mg Pt/ 2 cm ²	0.020- 0.030- 0.020 Electrolytic	31	25.4	23.3		Gave improved initial discharge voltage but deteriorated after 7 cycles.
15	-	"	-	"	0.020- 0.020- 0.020 Electrolytic	26.8	25.4	21.25	Following cells operated in horizontal position.	Cycled 7 times, showed deterioration with time.
16	-	"	-	"	0.020- 0.020- 0.020 Electrolytic	26.8	25.4	21.25	Same electrodes as above without washing.	Shows initial good performance but deteriorated with cycling.
17	-	"	-	"	Pure 0.060	28.0	25.4	26.5	After 5th cycle added water remtly. to improve performance.	Cycled 10 times Cell had low discharge voltage.
18	19	"	20	"	Pure 0.05G	22	25.4	32		Cycled 32 times 50 hours continuous operation. Exhibited slow deterioration with cycling. Final KOH 10.5%

TABLE 1 (con)

19	20 Mg. Pt. cm ²	20 Mg. Pt./ cm ²	Pure 0.050 F.C.	21.5	35	36.5	Cycled 2 times Cell flooded.
20	9 "	10 "	Pure 0.050 F.C.	22	40	35	Cell flooded.
21	10 Mg. Pt. 10 Mg. Pd.	20 Mg ₂ Pt. cm ²	Pure 0.050 F.C.	22	40	29	Cell cycled 3 times. Allowed to set overnight then cycled and found to show no deteriora- tion.
22	20 Mg. Pt. cm ²	20 Mg. Pt./ cm ²	Pure 0.050 F.C.	22	40	29	Cell used to check out new test set-up.
23	9 Mg. Pt./ cm ²	9 Mg. Pt./ cm ²	Pure 0.050 F.C.	22	40	29	Cycled 7 times. High charge voltage 1.7-1.9 volts. Discharge voltage degraded with time. Final KOH 33%
24	9 Mg. Pt./ cm ²	20 Mg. Pt./ cm ²	Pure 0.050 F.C.	22	40	29	Cycled 11 times High charge voltage 1.6-1.7 volts. Discharge degraded with time. Final KOH 37%
25	46 20 Mg. Pt./ cm ²	45 20 Mg. Pt./ cm ²	Pure 0.050 F.C.	22	40	29	Tested cell at 125°C Tested cell at high current discharge.

TABLE 1 (con.)

26	46	20 Mg.Pt./ cm ²	45	20 Mg.Pt./ cm ²	Pure 0.050	22	40.5	21	Used to check KOH loss. Final KOH 34.3%
27	46	20 Mg.Pt./ cm ²	45	20 Mg.Pt./ cm ²	"	24.0	40.5	31.5	Used 0.045 Spacer "Cell had pressure drop on first discharge
28	46	20 Mg.Pt./ cm ²	45	20 Mg.Pt./ cm ²	"	24.2	40.5	31.5	Cell improved capacity.
29	46	20 Mg.Pt./ cm ²	45	20 Mg.Pt./ cm ²	Pure 0.052	23	40.5	33	Cell improved capacity.
33		30 Mg.Pt./ cm ²		20 Mg.Pt./ cm ²	Pure 0.050	22	40.5	24	Cell used to test increased O ₂ catalyst Performance was not improved and test will be rerun
38		20 Mg.Pt./ cm ²		20 Mg.Pt./ cm ²	Pure 2X 0.030	26.5	40.5	36.4	Cell gave increased capacity but exhibited internal leakage.

this point causing gas leakage. (The design of the back-up plates, behind which are the gas storage chambers, consist of a 1/8 inch diameter hole which is drilled through the plate and a 0.060" deep cavity, approximately 1/2 inch in diameter underneath the electrode.) To alleviate this problem, screen spacers were placed under the electrodes at this point to prevent collapsing and to provide the desired matrix compression. This change was incorporated in cell number 6. However, upon cycling, it was found that non Faradaic pressure drops were also encountered with this cell due to internal, and/or external gas leakage. To eliminate internal leakage, it was decided to use a three layer matrix system of the same equivalent thickness as previous tests, i.e., 0.070".

Cell number 7 employed two layers of 0.020", and one layer of 0.030" electrolytic grade asbestos. This cell initially performed satisfactorily, and was cycled ten times continuously over a period of approximately 20 hours at which time the voltage on discharge fell below 0.7 of a volt and rose higher on charge. Figure 16 shows the typical voltage, total cell pressure, and differential pressure, during the charge and discharge of the cell. On experimenting with the cell, it appeared that the cell was capable of supplying 22 amp short duration current pulses at 0.8 volts. This indicates that performance degradation was probably due to concentration gradient build-up and concentration polarization within the cell. A small amount of water was added to the cell by supplying external gas and electrically discharging this gas into the cell to build up the water content in the mat. This addition of water improved the performance on both charge and discharge, and therefore it is reasoned that the difficulty was probably due to gas leakages during the course of the test which resulted in the drying out of the mat.

The next cell, number 8 was assembled with three layers of 0.020" asbestos. Compression was reduced from 7/4 ratio to a 6/4 ratio to evaluate the effect of improved ionic mobility on the reduction of concentration polarization. The electrodes used were the same as those used in test number 7, to show if this change would improve performance. No improvement in performance was obtained, and after 4 cycles, the test was discontinued.

An interesting observation made during the course of these initial single cell tests was the shift in differential pressure encountered when the cycle changed from charge to discharge. (Since these cells do not contain a volume balancing device, anomalous gas reactions are readily observable as pressure shifts during cyclic operation.) During the first few minutes of charge, a hydrogen gas differential pressure builds up. No oxygen gas is formed during this period, indicating an oxide formation on the cathode. This "stored" oxygen is later released during the first few minutes of discharge. The capacity that is stored varies somewhat from electrode to electrode, but in the cell geometry employed in these tests, a 6 to 10 psi differential was encountered. (See a differential change is shown in Figure 16 for cell number 7.) In addition, the oxygen storage process lowers the initial charging voltage and increases the initial discharge voltage. Subsequent testing showed that nickel oxide formation is responsible for the effect. Aside from possibly aggravating internal leakage, the differential pressure has not caused any thermochemical problems. Incorporation of a volume balancing device, such as is used in multi-cell operation, would eliminate pressure differentials.

The electrode back up plates used in the next series of tests were modified to include three radial grooves, 1/8" wide and 0.020" deep, to possibly improve gas access to the backside of the electrodes. Cell number 9 using electrolytic grade asbestos, was cycled 7 times (35 minutes discharge, 65 minutes charge) and left on open circuit over a 3-day week-end at 0 psig and 70°C. After this stand period, two

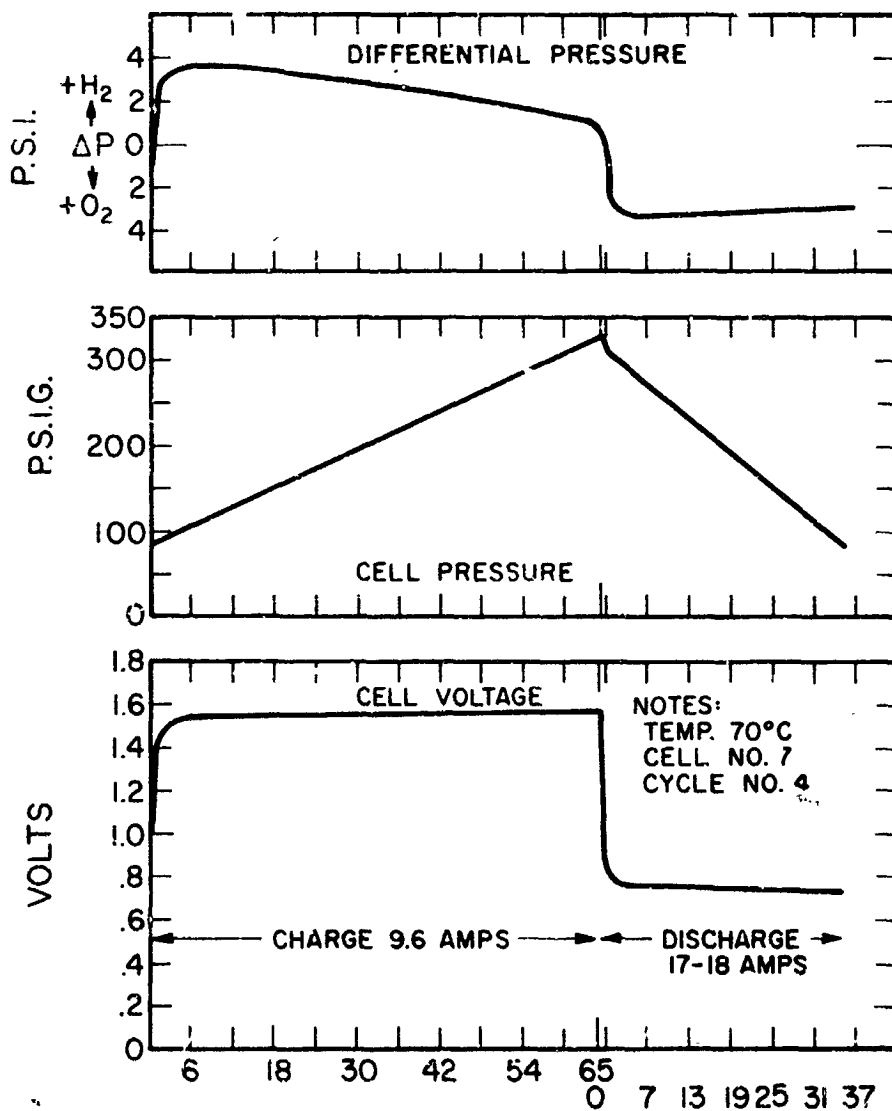


FIG. 16 CYCLING DATA - HYDROGEN-OXYGEN ELECTROLYTIC REGENERATIVE FUEL CELL

additional cycles showed that performance had deteriorated considerably, i.e., the discharge voltage was low, and the cell exhibited a high charging voltage. It was reasoned that possible deteriorations had taken place with the asbestos mat. Therefore, Cell No. 10 was assembled using a different grade of asbestos.

The mat used for Cell number 10 consisted of two layers of 0.035" thick commercial grade asbestos. In this configuration the cell exhibited considerable internal leakage and self-discharge. Therefore, the test was discontinued after a brief cycle period.

Cell number 11 consisted of two layers of the same material with inclusion of one layer of a cellulosic membrane type material (Visking) in an attempt to reduce internal cell leakage. The addition of the separator material resulted in increasing the cell internal resistance considerably. Consequently, the cell performed very poorly on both charge and discharge. It was then decided to go back to the mat configuration of cell number 7 which had given the best performance to date.

Cell number 12, using 3 layers of electrolytic grade asbestos, was cycled two times and left at temperature and 0 psig over a two day week-end. At the end of the period the cell exhibited degraded performance on both charge and discharge similar to cell number 9.

Cell number 13 was assembled using the same mat configuration as cell number 12 except the grade of asbestos was changed to fuel cell millboard, manufactured by the Johns-Mansville Company. Initial cycling of this cell indicated that the mat was low in water content. Water was added to the cell by pressurizing the cell and discharging the hydrogen and oxygen to form water within the cell. By doing this, it was possible to bring the cell performance up, but the cell still operated below the desired level. After 14 cycles, the cell was disassembled.

In order to improve the discharge voltage characteristics of the cell, it was decided to increase the catalyst loading in the electrodes. For cell number 14, an oxygen electrode containing 10 mg./cm² platinum catalyst, and 10 mg./cm² of palladium catalysts was used. The hydrogen electrode consisted of 20 mg./cm² of platinum. Initial performance of this cell was considerably better than all previous tests. Typical operating voltage on discharge with the original electrodes was approximately 0.7 volt. By increasing the catalyst loading, the cell operated at approximately 0.8 volt on discharge at the same current level. However, after cycling 7-8 times, this cell also deteriorated in performance.

The next cell number 15 was assembled to evaluate the effect of cell orientation, and was assembled and placed in the horizontal position. In addition, matrix compression ratio was reduced from a 7/4 ratio to a 6/4 ratio while slightly increasing the electrolyte/asbestos weight ratio. This cell was also cycled 7 times. Performance again deteriorated on both charge and discharge over this period. In order to determine if the deterioration was being caused by contamination of the electrodes, cell number 15 was rapidly disassembled and reassembled using the same electrodes without washing, but with a new asbestos matrix. This cell initially exhibited good performance, indicating that the electrodes had not been poisoned. After wet stand overnight, without being cycled, this cell also exhibited deteriorated performance.

In cell number 17, the grade of asbestos was changed to one layer of 0.060" fuel cell millboard. This cell was cycled 10 times and did not show any significant deterioration. However, the discharge performance was lower than some of the previous data obtained on initial cycling. Therefore cell number 18 was assembled containing a thinner (0.050") mat of the same type. The thickness was reduced in order to decrease the compression ratio. In addition, the quantity of electrolyte was increased. This cell was cycled two times then allowed to stand over night at temperature. It was then cycled

continuously for 30 cycles over a 50 hour period. Typical performance is shown on Figure 17. During the period of testing, there was some degradation in discharge voltage, and increase in charge voltage as shown in Figure 18.

In order to determine the cause of deterioration, the test was stopped and the gas compartments were flushed, reasoning possibly that impurities or gas mixing might have been the cause of the deteriorated performance. The cell was then put on discharge. Flushing did not improve the performance of the cell. Next an attempt was made to add water to the cell by pressurizing the cell and discharging the added gas into the mat. An increase in water content did not improve the cell performance. The cell was then disassembled, and the mat examined. As in previous tests, there was a discoloration of the mat adjacent to the hydrogen-electrode. Next, a sample of the mat was taken, and liquid KOH was squeezed out and titrated with 0.1 N HCl to determine the alkalinity content. This test showed that the liquid sample was approximately 10.5 percent KOH. Two separate samples titrated in the same manner gave the same result. (The cell had initially been assembled with 25.4 percent KOH.) Consumption of KOH such as was indicated, would explain the deteriorated performance.

In order to decrease the effect of KOH loss, it was decided to increase the electrolyte concentration from 25 percent KOH to 40 weight percent in future cell tests. Cells number 19 and 20 contained excessive quantities of electrolyte. When the cell was assembled, electrolyte was squeezed out of the asbestos mat into the individual gas cavities. On attempting to cycle these cells, it was found that they exhibited poor performance on discharge and extremely large, i.e., 10-20 psi differential pressures at the start of charge and discharge. The excessive differential pressures were attributed to flooding the electrode and back-up screen, forming N₂O which took part in the reaction on both charge and discharge. Cell number 21 was constructed with the electrolyte quantity reduced to 29 grams of 40 percent KOH. This cell

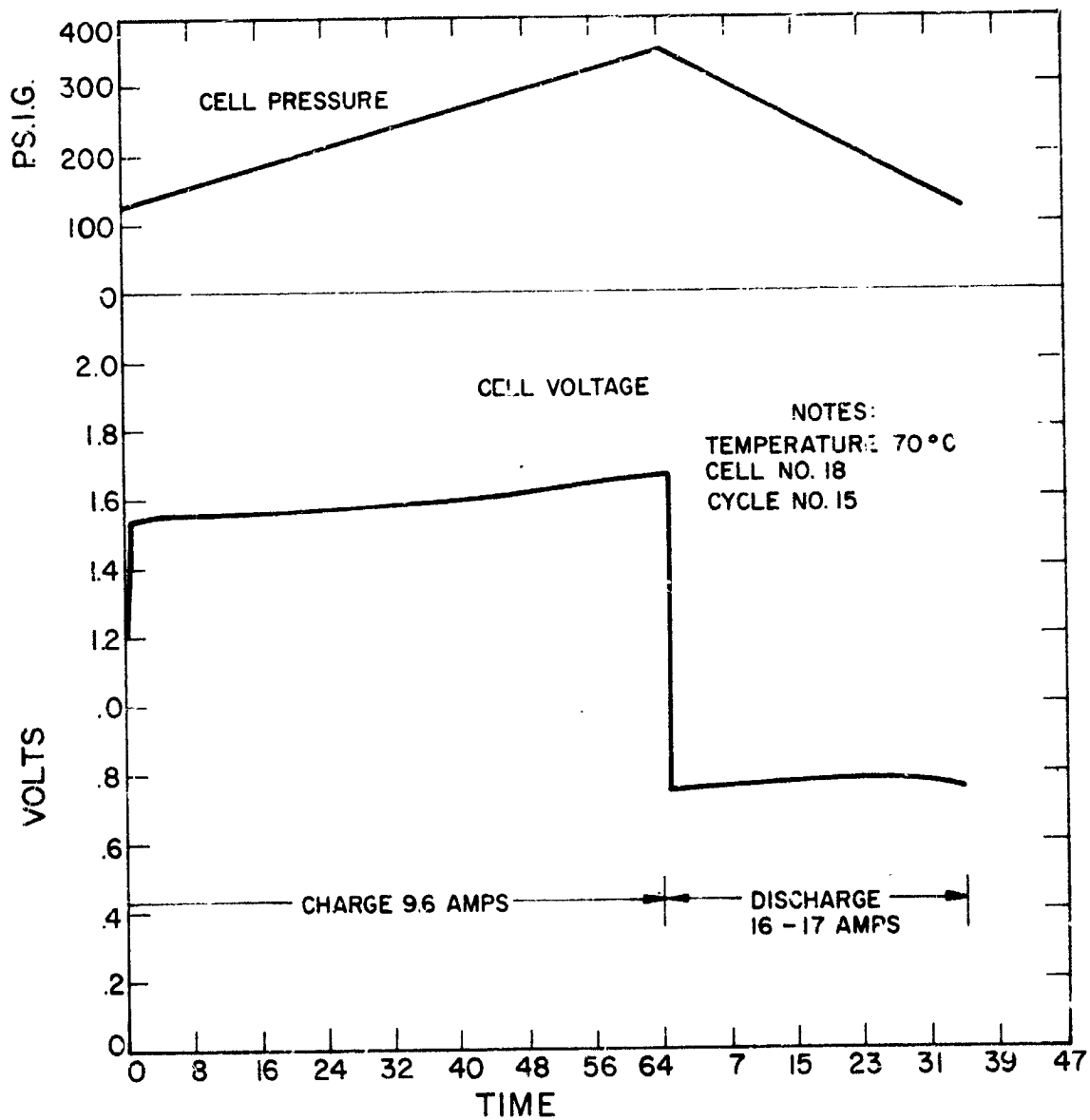


FIG. 17 CYCLING DATA - HYDROGEN-OXYGEN ELECTROLYTIC REGENERATIVE FUEL CELL

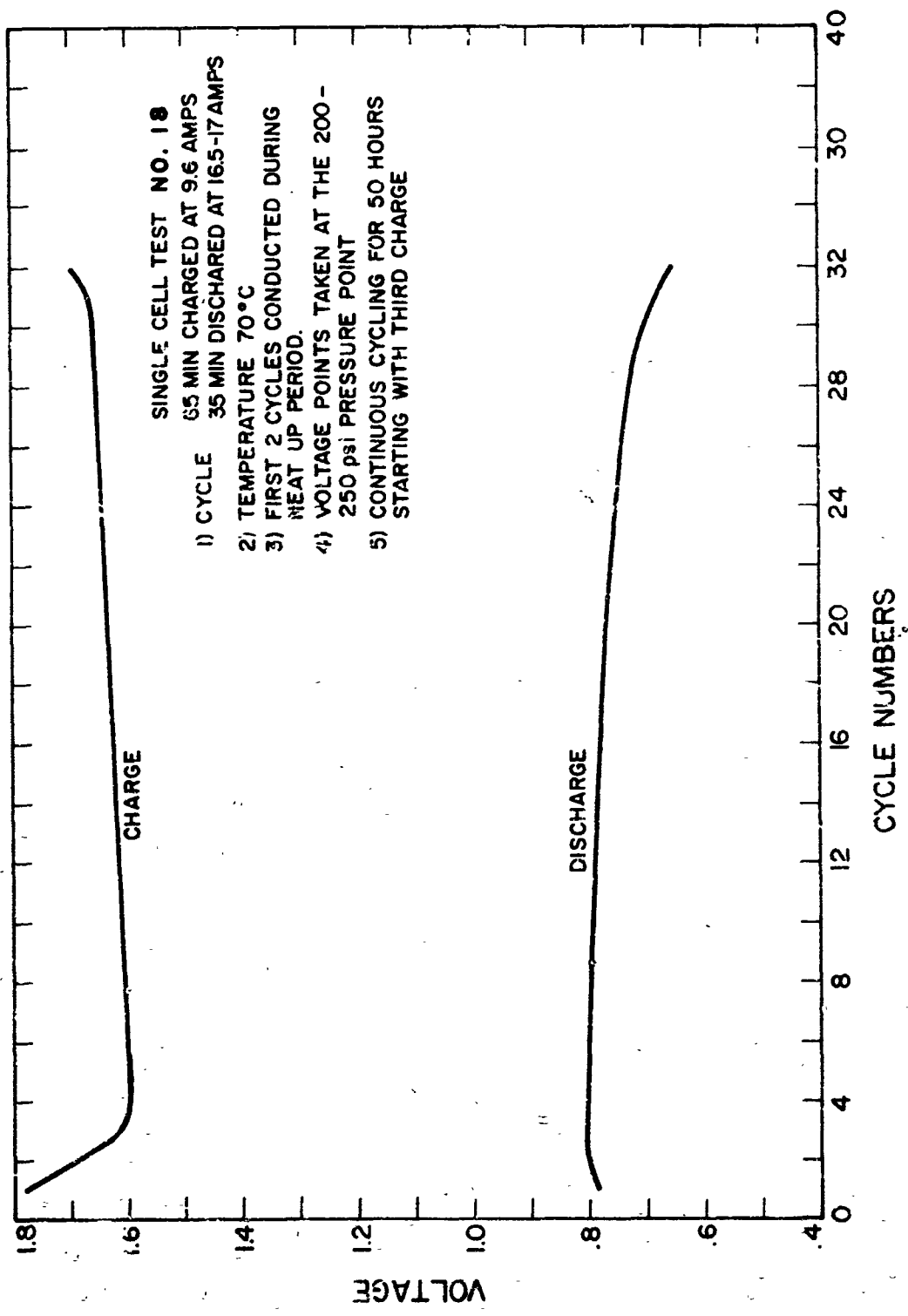


FIG. 18 EFFECT OF CYCLING ON CELL VOLTAGE

was cycled three times and allowed to sit in the open circuit condition overnight at 70°C, and then cycled a fourth time. The cell exhibited no obvious deterioration in performance during the period tested. It was decided to use this mat arrangement in initial tests with the 75 watt unit.

After completion of the first series of 75 watt cell tests, the single cell test oven was rearranged, and another series of single cells were assembled and tested. Cell number 22 was used to check out the new set up. Cell number 23 employed commercial platinized fuel cell electrodes. The initial discharge voltage at 16 amps was 0.8--0.82 volts, but the charge voltage ranged between 1.7-1.9 volts. After 6 cycles, the discharge voltage dropped to 0.76--0.78 volts. At this point the cell was disassembled and the electrolyte concentration in the mat measured. It was found to be 33 percent.

Cell number 24 utilized a Type AB-4 electrode on the oxygen side, and a standard EOS (20 mg/cm² platinum) hydrogen electrode. The discharge voltage at 16 amps initially was between 0.80 and 0.82 volts. The charge voltage ranged between 1.7 and 1.8 volts. After 11 cycles, the discharge voltage dropped to 0.78 volts. Again the cell was disassembled and the electrolyte concentration measured. This time it had also dropped from 40 percent to 33 percent. Figure 19 shows the E-T data using commercial electrodes.

Cell number 25 was assembled to test capabilities of the cell at elevated temperatures. Figure 20 shows E-I data for the cell at 125°C. The data obtained at 125°C is quite good, in that currents approaching 100 amperes were obtained at 0.55 volts including an IR drop of \approx 0.2 volt. Due to overheating of the load bank and associated electrical hardware, the high current levels, i.e., >50 amps were only measured for periods of time less than 30 seconds. However, the values obtained are indicative of the high pulse power the unit is capable of at slightly elevated temperature. Testing of this cell was discontinued in order to use the instrumentation for a second test series with the 6-cell, 75 watt unit.

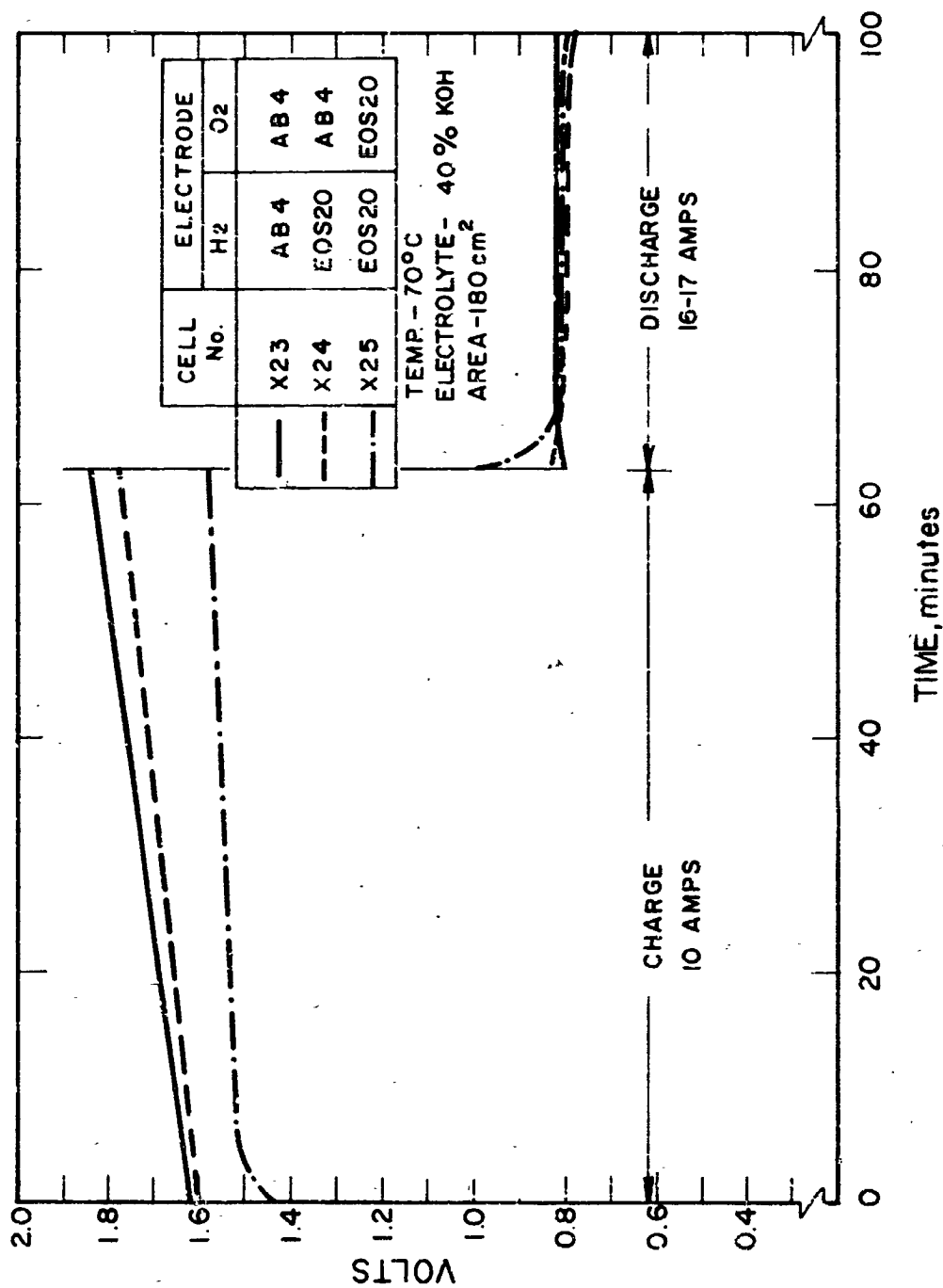


FIG. 19 COMPARISON BETWEEN CONFLUCIAL AND EOS ELECTRODES

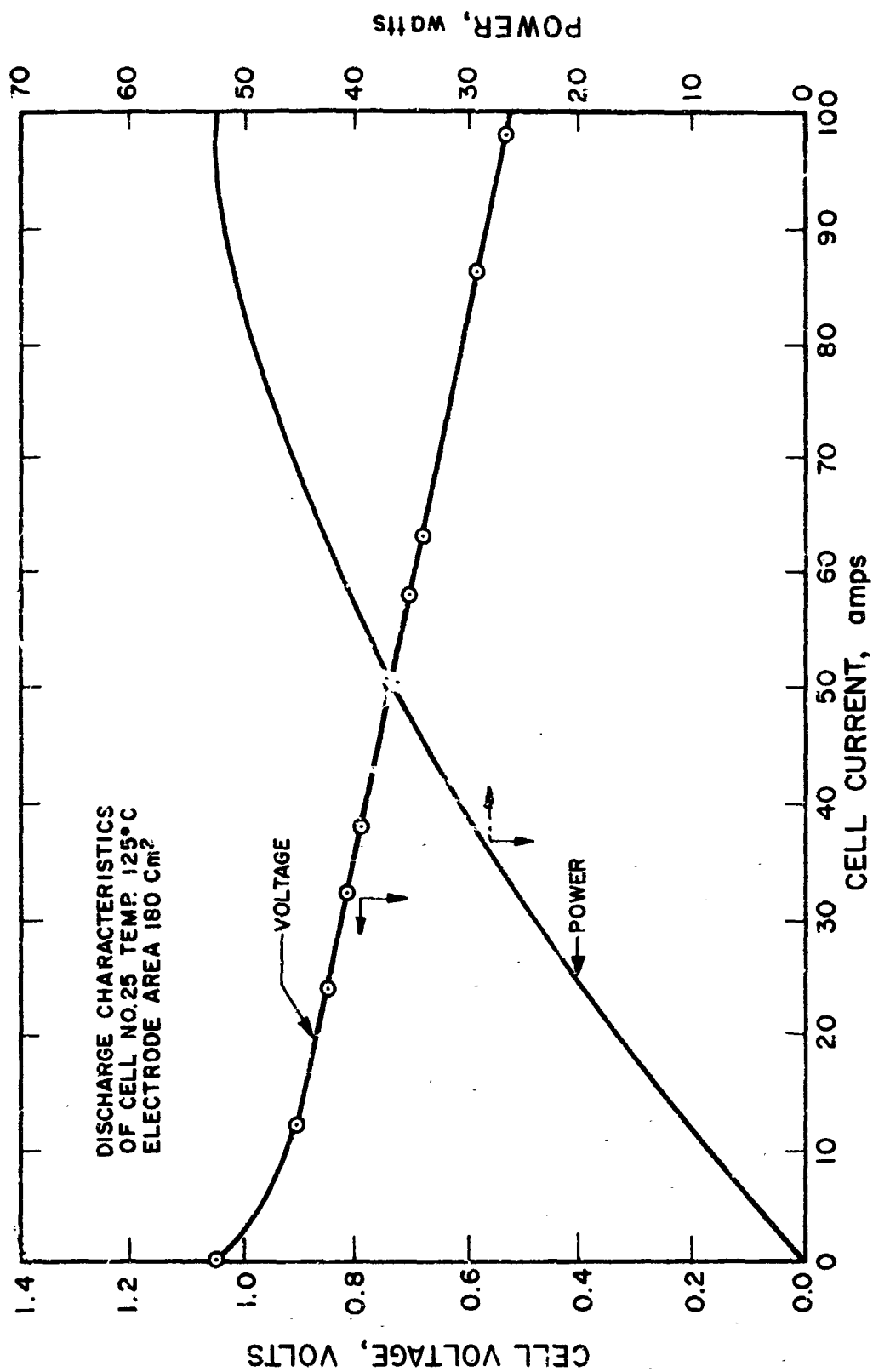


FIG. 20 CELL NO. 25 DISCHARGE CHARACTERISTICS

Cell number 26 was assembled to demonstrate single cell performance to the NASA technical monitors of the program, who visited EOS during this period. This cell was cycled 2 times, and subjected to discharges at various currents from 10-91.5 amps for short periods of time. The cell was then allowed to sit overnight at the elevated temperature, after which, the mat was removed, and the KOH concentration analyzed. It was found to have dropped from 40 to 34 percent KOH. This continued change in KOH concentration as a function of time was and is cause for concern. A series of tests, discussed in detail in the next section, were then initiated to determine the reason for this loss of KOH.

Phase II of the program calls for a 500-watt unit with individual cells of 6-inch diameter having a capacity of 21 amp. hours. Prior to cell number 27, all work has been done with the 6-inch diameter cell of nominal 10 amp. hour capacity. In order to increase capacity, mat composition compression, dry-weight, and wet weight must be modified in order to contain sufficient water to provide 21 amp. hours capacity. A series of cell tests was initiated to study mat variations that could be used to improve this capacity. Cell number 27, the first in the series, consisted of a thicker mat containing a quantity of electrolyte above what has been previously used. In order to obtain the higher capacity using the fixed volume single cell, the cell was electrolytically charged to 500 psig instead of the normal 350-400 psig. When the first discharge was started, a rapid pressure drop was noted. The test was then stopped, the cell vented, disassembled and examined. The asbestos mat periphery was distorted and there was a black deposit in the gas compartments. (Note, the mat edges were not rubberized.) It was decided that the mat had possibly been placed off center, and therefore cell number 28 was assembled using the same mat arrangement.

The mat configuration of cell number 28 provided increased capacity, but did not provide the full 21 amp hours desired. Voltage rose high at the end of charge indicating drying out of the mat.

Therefore, initial discharge voltage was quite low, and at the end of discharge, voltage fell off rapidly. This performance is shown in Figure 21.

Cell number 29 contained a mat with reduced compression and increased electrolyte. The cell was cycled at charge currents of 10 and 20 amps. and a discharge current of 20 amps. The cell charged well and discharged well except at the end of discharge when performance fell off rapidly, as shown in Figure 22. Poor Faradaic efficiencies were noted. To improve discharge performance and to avoid the apparent flooding at the end of discharge, 100 psi gas equivalent was vented to reduce the water content at the end of the fourth cycle. At the start of the fifth cycle discharge, the pressure dropped rapidly, i.e., 200 psid. The test was then stopped, the cell vented, disassembled and examined. Once again the edges of the mat were badly distorted and black deposits were found in the gas compartments. In these tests the mat compression had been reduced, and apparently the compression on the mat edges was not sufficient to prevent gas leakage without an added edge sealant. A mat edge sealant, viton rubber, had been used in most previous multi and single cell tests. However, as discussed in the next section, this sealant reacts chemically with KOH. A new material, an ethylene-propylene rubber has been recently evaluated and found to be satisfactory. Using this sealant will eliminate the edge seal problem.

To further increase cell capacity to the desired 21.4 amp. hrs., additional variations in mat thickness, compression, and electrolyte quantity have been studied. Cell number 38 consisted of a mat of two layers of 30 mil fuel cell millboard asbestos which was impregnated with 36.4 grams of 40 percent electrolyte and placed within the cell. A 0.040" spacer was used in the periphery of the cell, but the 10 mil gas distribution screens behind the electrodes were removed. Therefore, there was a 60 mil space between the electrodes, but the outer edge of the mat was compressed between the tank halves to 0.040". The cell was cycled and charged to 500 psig and then discharged. This arrangement provided

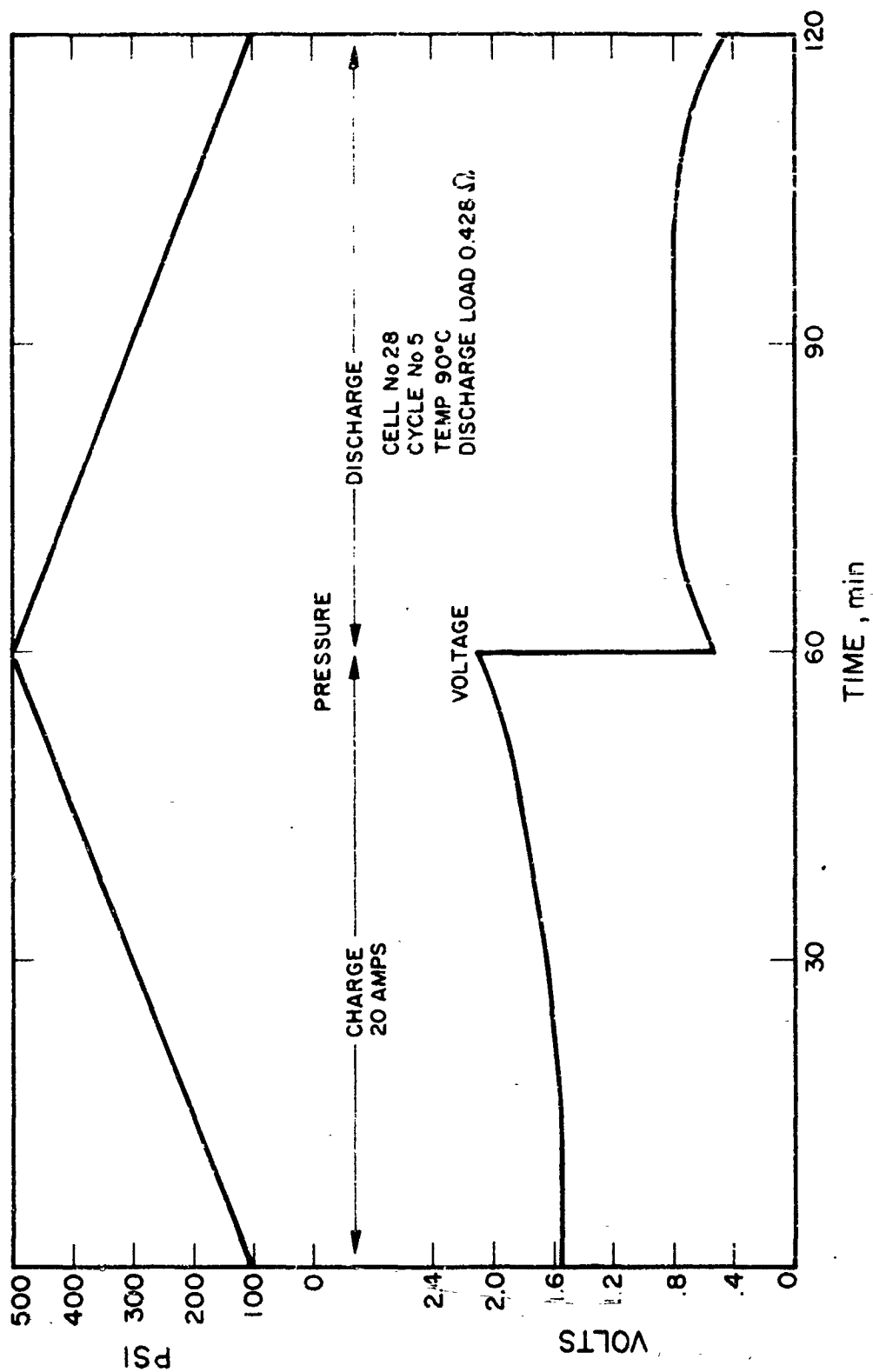


FIG. 21 INCREASED CAPACITY TEST, CELL NO. 28

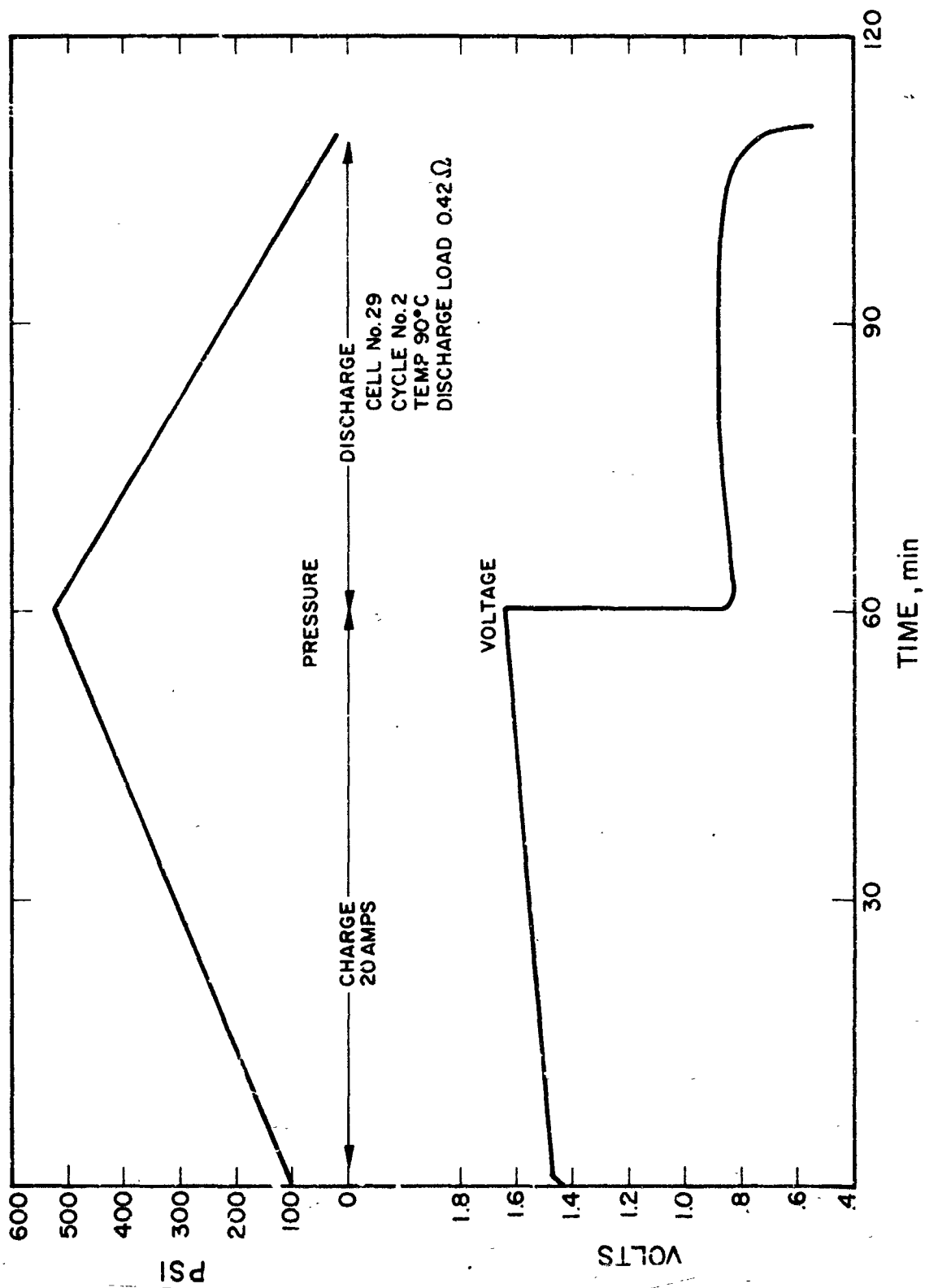


FIG. 22 INCREASED CAPACITY TEST, CELL NO. 29

increased cell capacity, but still did not achieve the 21.4 amp. hrs. desired. Figure 23 shows typical performance of this cell.

Table II shows a comparison of all high capacity tests thus far. As can be seen, cell number 43 has given the best results yielding 20.5 A.H. This cell had no initial voltage dip, and only during the last 10 minutes of discharge did the performance fall off. Some additional tests are planned to determine the correct combination of asbestos thickness, compression and water content to achieve the optimum capacity and performance.

3.8 KOH Consumption Investigation

In order to determine the cause of KOH consumption during the operation of cells, a series of tests was conducted to determine if this consumption is caused by any impurities within the actual electrodes. Each cell was assembled late in the afternoon and allowed to come up to temperature (70°C) overnight. The cell was cycled the following day until late afternoon, and then disassembled, and the mat KOH sample was analyzed. The variables in the test consisted only of the method of preparation of the electrodes. The variables are listed in Table III which also shows the results obtained in the analysis. As can be seen the electrolyte concentration in all cases dropped to 30-35 percent KOH. No effect could be attributed to the electrodes. To compare this finding with direct chemical tests, fuel cell grade asbestos mats were soaked with the same quantity of electrolyte that is normally used in the cell test. These were stored at room temperature and at 70°C, and then analyses of KOH concentration were made on these mats. The results are shown on Table IV.

During both single cell and multi-cell tests in which the asbestos mat had been rubberized with viton on the periphery, there were definite visual indications of chemical reaction between the viton and the electrolyte. Further, viton "O" rings used initially in the single cell were severely attacked by the same electrolyte. These visual indications, plus test results showing that noticeable changes in

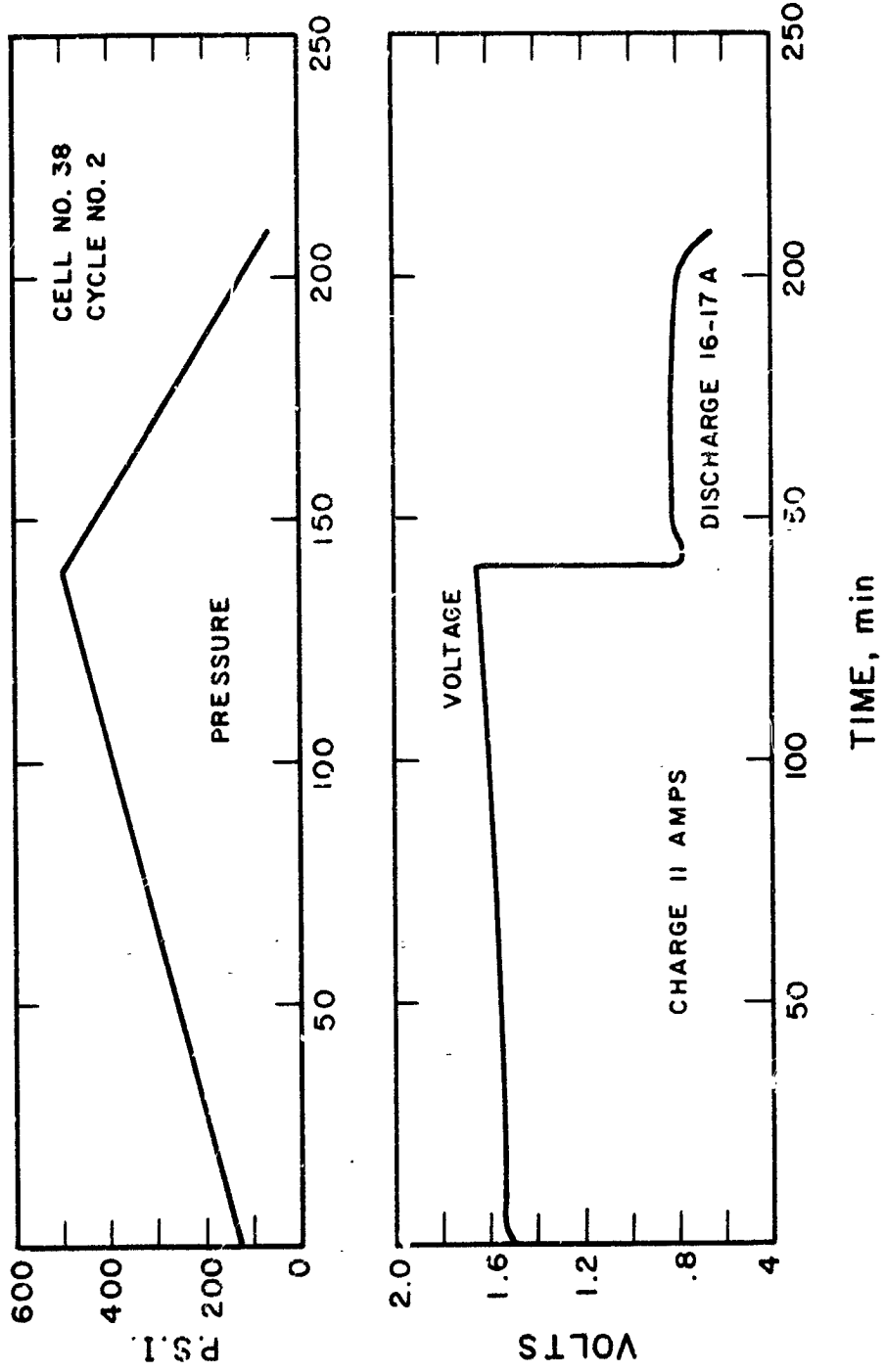


FIG. 23 INCREASED CAPACITY TEST, CELL NO. 38

TABLE II SINGLE CELL HIGH CAPACITY TESTS

Cell No.	Mat Thickness Inches	Dry Wt. Gms.	Electrodes Spacing	KOH			To End of Charge		To .7 Volt Cut Off			Avg. Curr. Amps	Comments
				%	Gms.	Wt.	PSI	Volts	Time Min.	PSI	A.H.		
102	.050	22	.050	40.5	29		450	1.65	57	100	14.2	15	From 75W unit voltage/cell calculated
27	.055	24	.045	40.5	31.5								Cell had sharp pressure drop.
28	.055	24.2	.045	40.5	31.5		485	2.07	50	140	15	18	Had initial dip in voltage
29	.052	23	.045	40.5	33		520	1.64	50	140	16	19.2	Had voltage fall off at end of discharge.
38	2 x .030	26.5	.050	40.5	36.4		500	1.66	69	70	18.9	16.5	On 3rd cycle cell had internal leakage.
40	2 x .030	27.2	.060	40.5	36.4								.001 layer permion had high internal resistance.
41	2 x .030	26.6	.060	40.5	36.4		500	1.90	56	145	16.1	17.3	Large initial dip Excess liquid in cell.
42	2 x .030	26.7	.060	40.5	34.4		500	1.77	71	63	19.2	16.4	Initial dip in voltage.
43	1 x .060	28.1	.060	40.5	35		500	1.595	72+	70	20.5	17.3	Cell performed well except voltage fell off at end of discharge.

TABLE III

KOH LOSS IN SINGLE CELL TESTS WITH VARIABLE ELECTRODE TREATMENTS

Cell No.	O ₂ Electrode No.	H ₂ Electrode No.	Electrode Treatment	Initial KOH %	Final KOH %
30	58	57	None	40.5	32.1, 32.3
31	60	59	None	40.5	32.7, 32.8
32	58	57	Cell No. 30 electrodes washed with hot distilled water	40.5	33.1, 31.4
34	60	59	Cell No. 31 electrodes washed with hot distilled water	40.5	32.6, 32.3, 34.6
35	55	56	After processing washed with hot KOH and then washed with hot distilled water.	40.5	33.8, 32.4
36	53	52	"	40.5	29.9, 29.6, 33.2
37	53	52	Cell No. 36 electrodes washed with hot distilled water.	40.5	35.1, 35
39	55	56	Cell No. 35 electrodes washed with hot distilled water.	40.5	36.7, 33.1, 32.9

Note: All cells used 0.050 thick 22 gm fuel cell asbestos, KOH 29 gms. electrodes catalyst 20 Mg.Pt./cm²

TABLE IV
KOH ANALYSIS TESTS

Test Sample	Storage Period	Storage Temperature	Original KOH %	Final KOH %
Fuel Cell Asbestos	136 hr.	R.T.	40.5	35.7, 35.6
"	136 hr.	R.T.	40.5	35.1, 35.5
"	88 hr.	70°C	40.5	33.6, 33.4
"	88 hr.	70°C	40.5	34.6, 34.6
Fuel Cell Asbestos with viton rubber edging	136 hr.	R.T.	40.5	35.5, 35.8
"	136 hr.	R. T.	40.5	34.6, 34.7
"	88 hr.	70°C	40.5	32.9, 33.2
"	88 hr.	70°C	40.5	33.2, 33.7

electrolyte concentration, i.e. >10 percent only occurred when the asbestos was rubberized with viton, are strong signs that the rubber reaction contributed to the KOH degradation. However, tests made in a similar manner to that described above, but including rubberized edges, showed no significant difference, as is shown on Table IV. This result is somewhat disconcerting, in view of the obvious visual deterioration of the rubber. There are two possible explanations for the results obtained, namely (a) the period of time allowed for the reaction, i.e., 88 hours was insufficient or (b) the test procedure was unsatisfactory. The latter explanation has been investigated in some detail. Titration solutions have been checked and rechecked, initial electrolyte concentrations checked and rechecked, multiple samples analyzed to check for reproducibility, and test samples double sealed to prevent water loss. As a result of this checking, we feel that the former explanation is the most reasonable answer.

Regardless, it has been established that KOH slowly reacts with the asbestos or impurities therein. This reaction apparently results in a 6-10 percent reduction in concentration in a period of 24-48 hours. However, it remains to subject cells to long periods of cycling to assure that the reaction does not proceed beyond this level. If the KOH concentration stabilizes above 30 percent, there is no problem.

3.9 Preliminary 500 Watt Cell Design Weights

As a result of the recent series of tests indicating the high performance achievable with the new cell design and improved electrodes, preliminary weight calculations of a 500 watt, 35 cell unit were undertaken. Two sets of calculations were made, one using existing components, and the second using improved lower weight components which we feel are well within the current state of the art. Table V shows the itemized component weights used in the calculated cell weights. Further weight reductions are possible, using advanced materials and improved designs. Details of such reductions are given in the next section.

		35 Cell Unit		35 Cell Improved Design	
		<u>Existing Design Wt.</u>		<u>Estimated Achievable Wt.</u>	
		(gms)		(gms)	
		<u>Each</u>	<u>Total</u>	<u>Each</u>	<u>Total</u>
1.	Separator Plate	159.5	5750	140	5040
2.	Insulating Spacers	21.9	767	20	700
3.	Electrodes	21.0	1470	15	1050
4.	Asbestos Mat	22.0	770	21	735
5.	Gas Distribution Screens	12.1	846	0	0
6.	Pressure Balancing Bellows	1328	1328	800	800
7.	K ₂ end plate	1139	1139	360	360
8.	O ₂ end plate	1375	1375	480	480
9.	Stack bolt assembly	65	780	65	780
10.	Bellows "O" ring	20	20	15	15
11.	Flange "O" ring	28.1	56	20	40
12.	Flange Bolt Assembly	31.5	378	30	360
13.	Electrical connector	200	200	200	200
14.	Electrolyte	29	960	26	860
15.	Tankage	6400	6400	5270	5270
		22,390 gms.		16,690 gms.	
		or		or	
		49.3 lbs.		36.5 lbs.	

How Weight Reduction
Accomplished

Additional gas grooves.
Use epoxy instead of teflon.
Use Ac electrode on O₂
Use .0.050" instead of 0.054"
Eliminate Screens

Use thinner flange.
Use Mg instead of Al and reduce to 1/4".
Use Mg instead of Al and reduce to 1/4"
No change.
Reduce thickness.
Reduce thickness.
Use smaller washers.
No change.
Reduce KOH to 30 percent.
Reduce flange diameter and use 50K psi alloy.

TABLE V. COMPONENT WEIGHTS FOR 500 W 600 W.H. MODEL

3.9.1 Potential Improvements in Regenerative Fuel Cell Design Using Advanced Light Weight Materials

The purpose of this discussion is to review the results of a preliminary analysis which was performed to determine the potential reduction in weight of a regenerative fuel cell which may be possible by utilizing advanced light weight materials. A review of the existing design concept scaled up to the 500 watt nominal power rating indicates that with relatively few and simple improvements, the weight of such a system could be reduced to the 35-40 lb. level. Such a unit would utilize magnesium cell separators, and 6061-T6 aluminum alloy gas tankage. The unit would be capable of an energy capacity of 600 watt hours at a 500 psig peak storage pressure, at a temperature of 100°C and power drains of either 500 watts or 1000 watts. The cell stack assembly would have an estimated weight of about 20-25 lbs., and the gas tankage assembly would have an estimated weight of about 10-15 lbs. The best estimate of the energy to weight ratio of such a unit would be 15-17 watt hours per lb. The unit would be essentially identical to the present design with minor modifications to reduce weight in non-critical areas. The principle change in operation would be achieved by raising the operating temperature to the 100°C level to allow a drain of 1000 watts at the short duration discharge.

To examine the feasibility of further reductions in weight, several advanced materials were considered for use in the present design. The existing cell stack assembly uses nickel plated magnesium separators with a density of about 0.065 lbs. per cubic inch. If one of the new light weight aircraft structural materials such as the magnesium-lithium alloy LA141A can be used, with a density of 0.049 lbs. per cubic inch, an immediate reduction of the cell stack of about 4-5 lbs. is possible. The use of this and other similar magnesium-lithium alloys is gaining rapidly, and in this particular application would

depend on the development of a suitable nickel plating process for protection against electrochemical attack. Other design improvements in the cell stack bolts and end caps are certainly possible with improved high strength, light weight materials, thereby making another 2-3 lb. reduction in weight feasible. Therefore, the minimum weight of a 600 watt hour capacity cell stack is probably on the order of 16-18 lbs., as compared to the present average of about 25 lbs.

The gas tankage is the other most critical weight element of the system. The existing tank design for a 500 watt unit would utilize the 6061-T6 aluminum alloy with a working yield stress of 33,000 psi, and a density of 0.1 lbs./cu. inches. There appear to be several alternative materials which could make significant reductions in the present tank configuration, which appears to weigh about 12-15 lbs. If one of the structural titanium alloys could be nickel plated, or coated with an inert material such as teflon, to protect against spontaneous reaction in the pure oxygen environment, then a design working stress of 100,000 psi can be used with a material density of 0.160 lbs./cu in.; thereby resulting in a tank wall thickness of approximately 0.030". The weight of the gas storage tankage utilizing titanium would therefore be about 7-8 lbs. including flanges.

As another approach, if one of the new high strength weldable aluminum alloys such as Kaiser 7039, or Alcoa X-7106 proves to be satisfactory at operating temperatures of 250°F, then a design yield stress of 45,000 psi would be available. These materials can be expected to have all of the desirable properties of the older aluminum alloys, and would result in a gas tankage weight of 10 lbs. Another alternative, and probably the most attractive, considers the use of the new beryllium-aluminum alloys developed by Lockheed Missiles and Space, and recently licensed to the Brush Beryllium Company. These alloys vary in percentage from 25-45 weight percent

of aluminum with the remainder pure beryllium. They appear to have good ductility and corrosion resistance, and also are sufficiently weldable to be used for tank design. The main advantage of these alloys is a working yield stress of 70,000 psi with a density of 0.074 lbs./cu in. If this material becomes available and can be used in this application, it would result in a total tank weight of 6 lbs. including flanges. The last alternative considered, involves the use of glass filament wound tanks. Although the present technology has resulted in very high strength to weight ratio configurations, it is not believed that these tanks are practical for this application at this time. The two factors which influence this conclusion are the unpredictable effect of temperature cycling on the strength characteristics of the glass filament structure, and secondly, the heat transfer requirements of the fuel cell assembly whereby a high thermal conductivity, both axially and radially, on the tank structure is desirable for heat rejection purposes. However, it is likely that with current developments underway, a high temperature metallic filament wound tank will become feasible for this application in the near future, utilizing some of the high temperature resins and adhesives presently being developed.

The conclusions derived from this preliminary study of trends in performance are significant. The minimum weight of a 600 watt hour regenerative fuel cell assembly using the most practical advanced structural materials which could become available in the next 2-3 years, appears to be a lower limit of about 20 lbs., and a likely weight of about 25 lbs. This would result in a system energy to weight ratio of 24 watt hours per lb. If the capacity of the existing cell stack could be increased by improving the water storage capability, the energy to weight ratio of the system could probably be raised to at least 30 watt hours per lb. This appears to be a reasonable goal for a regenerative fuel cell battery operating at 100°C after a development prototype has been completed and

reliable cyclic operation is achieved. For the foreseeable future, this system appears to be significantly superior to other secondary batteries on an energy/weight basis.

4. CONCLUSIONS

The major test objective of the Phase I portion of the program, i.e., continuous operation of a 6 cell regenerative unit for 48 hours at > 75 watt has been achieved. In addition, large improvements in performance have been achieved with single cell tests, indicating higher performance levels can be achieved with multi-cell units. Difficulties encountered in electrolyte consumption appear to have been resolved. Preliminary designs for a 500 watt, 600 watt-hour unit indicate that 15 watt hours per pound can be achieved. A 20 watt hour per pound unit is attainable if the use of certain high strength to weight materials can be utilized.

5. PLANS FOR THE NEXT QUARTER

Having completed Phase I, major emphasis will be shifted to designing and fabricating a 500 watt, 600 wat-hour unit. In order to achieve the performance required, an additional single and six cell test series will be required. This series will include tests to (a) optimize asbestos mat geometry and electrolyte content, (b) evaluate long term cycle effects (c) increase catalyst loading and (d) self discharge effects.

It is anticipated that the design of the first 500 watt unit will be completed by January 31, 1965. After design approval has been obtained, the various components will be fabricated. Initial assembly of the first 500 watt unit should occur early in the second quarter of 1965.

REFERENCES

1. EOS, 2nd Quarterly Report-4110-2Q-1, 26 July, 1964,
NAS Contract 2-3781.